Macroscopic QED in Linearly Responding Media and a Lorentz-Force Approach to Dispersion Forces

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It is nice to know that the computer understands the problem. But I would like to understand it too.

Eugene Wigner

Chapter 1

Introduction

It is well-known that polarizable particles and macroscopic bodies (i.e., matter whose electromagnetic properties are described terms of macroscopic state variables) are subject to forces in the presence of electromagnetic fields. This may be the case even if the fields vanish on average and the bodies do not carry any excess charges and are unpolarized, because of fluctuations. In classical electrodynamics, fluctuations may be thought of as resulting from 'ignorance': it is only the lack of precise knowledge of the state of the sources of a field that makes one resort to a probabilistic description. Classical fields can therefore be non-fluctuating as a matter of principle, which is the case if the sources, and thus the field, can be regarded as being under strict, deterministic control. Specifically, the classical electromagnetic vacuum (having no sources whatsoever) does of course not fluctuate—all moments of the electric and induction fields vanish identically, which implies the absence of any interaction with matter.

In quantum electrodynamics, the situation is rather different, since fluctuations are present in general even if complete knowledge of the quantum state is assumed to be available. Since (genuine) joint probability distributions cannot be introduced for the non-commuting, operator-valued field quantities, a strictly non-probabilistic regime (that is to say, a δ-function-like joint distribution) does not exist either. Hence non-vanishing moments occur inevitably—at least some of the field quantities fluctuate whatever the quantum state. In particular, fluctuations are present also if the field–matter system can be assumed to be in its ground state (vacuum), where only quantum fluctuations are responsible for the forces exerted on the matter that interacts with the field. In this case, it is common to speak of vacuum forces or dispersion forces, which obviously represent a genuine quantum effect. A renewed interest in the dispersion forces has emerged over the last years, partly stimulated by the progress in the fabrication and operation of nano-mechanical devices, where dispersion forces play an ambivalent role. Despite being vital for the design and operation of such devices, they may on the other hand lead to their destruction (see, e.g., Refs. [1–4]). Together with a number of other observable effects that can be attributed to the interaction of the fluctuating electromagnetic vacuum with material systems (such as spontaneous emission or the Lamb shift), the experimental demonstration of dispersion forces has been widely regarded as constituting a confirmation of quantum theory [5].

On the microscopic level, a well-known dispersion force is the attractive van der Waals (vdW) force between two unpolarized ground-state atoms, which can be regarded as the force between electric dipoles that are induced by the fluctuating vacuum field. In the non-retarded (i.e., short-distance) limit, the potential associated with the force has been first calculated by London [6, 7]. The theory has later been extended by Casimir and Polder [8] to allow for larger separations, where retardation effects cannot be disregarded. Examples of dispersion forces on macroscopic levels are the force that an (unpolarized) atom experiences in the presence of macroscopic (unpolarized) bodies—referred to as the Casimir–Polder (CP) force in the following—and the Casimir force between macroscopic (unpolarized) bodies (for reviews see, for example, [5, 9]). Since macroscopic bodies consist of a huge number of atoms, both the CP force and the Casimir force can be regarded as macroscopic manifestations of microscopic vdW forces, and both types of forces are intimately related to each other. They cannot be obtained, however, from a simple superposition of two-atom vdW forces in general, since such a procedure would completely ignore the interaction between the constituent atoms of the bodies, and thus also their collective influence on the structure of the body-assisted electromagnetic field [10].

Although it is certainly possible, in principle, to calculate CP and Casimir forces within the framework of microscopic quantum electrodynamics (by solving the respective many-particle problem in some approximation), a macroscopic characterization of the bodies involved is preferable in general. The reason is that even if a fully microscopic, *ab initio* theory of the dispersion forces were given and explored to its conclusions, it would ultimately be necessary to relate the necessarily huge number of microscopic parameters involved (such as coupling constants) to a small number of macroscopic, experimentally accessible quantities. In fact, the macroscopic bodies involved in dispersion-force experiments are in practice always characterized in the manner familiar from the macroscopic electrodynamics of continuous media (i.e., in terms of macroscopic constitutive relations and/or boundary conditions), which is therefore a suitable language to formulate the problem. One may clearly restrict attention to linear media when discussing dispersion forces.

Over the decades, different macroscopic concepts to calculate the CP and Casimir forces have been developed, but compared to the large body of work in this field, not too much attention has been paid to their common origin and consequential relations between them (see, e.g., Refs. [11–14] and [R4]). Moreover, the studies have typically been based on specific geometries such as simple planar structures, and weakly polarizable matter has been considered. More attention has been paid to the relations between Casimir forces and vdW forces, but again for specific geometries and weakly polarizable matter (see, e.g., Refs. [8,10,11,15–18]). Relations between CP forces and vdW forces have on the other hand been established, on the basis of both microscopic and macroscopic descriptions, and, moreover, without the assumption of weakly polarizable matter [19–21]. These relations show clearly that the CP force acting on an atom in the presence of a dielectric body whose permittivity is of Clausius–Mossotti type can be regarded as being the sum of all the many-atom vdW forces with respect to the atoms of the body. It is thus only natural to ask if the connections between the CP force and the Casimir force may be understood in a similar way and expressed in general terms. One aim of this work is to provide answers to this and related questions.

Any satisfactory macroscopic theory of dispersion forces should of course be based on a consistent quantum theory of the macroscopic electromagnetic field in the presence of media. Unfortunately, many accounts of CP and/or Casimir forces found in the literature have to be criticized in this regard. A typical example is the calculation of the Casimir force between macroscopic bodies within the so-called mode summation approach (which is close to Casimir's ideas), where one assigns some geometry-dependent [distance parameter(s) d] electromagnetic vacuum energy

$$E(d) = \tfrac{1}{2}\sum_m \hbar\omega_m(d) - \tfrac{1}{2}\sum_m \hbar\omega_m(d\to\infty) \tag{1.1}$$

to the body-assisted field, and regards it (with suitable regularization) as the potential of the Casimir force. The geometry-dependent mode frequencies $\omega_m(d)$ needed in Eq. (1.1) are defined by an eigenvalue problem obtained from (the source-free version of) Maxwell's equations in the presence of the macroscopic bodies (see, e.g., Ref. [5]). The calculations that follow this route are usually based on a quantization scheme where the electromagnetic field is expanded in modes (obtained from the mentioned eigenvalue problem), and quantized in analogy with the well-known method of quantizing the field in free space. Within such an approach, unitarity demands that the modes be genuine normal modes with real mode frequencies which is, however, the case only if the material bodies are represented in a comparatively crude way, e.g., by perfect-conductor boundary conditions, or as non-dispersing and non-absorbing dielectrics. On the other hand, force calculations on the basis of Eq. (1.1) or equivalent expressions have been put forward in the literature also in cases where a more advanced description of the bodies (featuring dispersion and absorption) is considered. The eigenvalue problem used to determine the mode frequencies then exhibits several non-standard features, so that the corresponding (non-normal) mode formalism tends to become somewhat heuristic. Attempts to justify (formally) the use of Eq. (1.1) even in these cases typically proceed by rewriting it as a complex contour integral which is then argued to have a wider range of applicability than Eq. (1.1) itself, or by invoking a fictitious auxiliary system to define the modes (see, e.g., Refs. [5, 22] and references therein). The formal arguments effectively involve the analytical continuation of an (in general non-Hermitian) eigenvalue problem with respect to a parameter (the frequency) and may thus be delicate mathematically. [One possible mathematical complication is related to so-called spectral singularities, which are points in parameter space where the (in general bi-orthogonal) eigenfunctions of a parameter-dependent eigenvalue problem are not complete. This can happen even at points where the eigenvalue equation depends on the parameter analytically.] The possibility of a formal generalization of Eq. (1.1) from non-dispersing and non-absorbing to dispersing and absorbing media may hence be doubted already on purely formal grounds. Aside from the mathematical problems, the more important problem is that the physical meaning of Eq. (1.1) is far from being transparent when dispersion and absorption are taken into account. In essence, Eq. (1.1) is equivalent at best to an energy-like expression whose meaning is physically questionable as soon as the field inside a medium is considered (see below).

A different and physically much more transparent approach to the calculation of dispersion forces is based on the so-called Rytov-Lifshitz fluctuation electrodynamics, which has first been used by Lifshitz to calculate the Casimir stress between two dispersing and absorbing dielectric half-spaces separated by an empty interspace [10]. To find the force on one of the half-spaces, only the stress tensor in the free-space region between the half-spaces is required. The question as to how the Casimir force between bodies should be calculated if the interspace between them is not empty arises quite naturally. Frequently, expressions that seem reasonable at first glance—such as Minkowski's stress tensor—have been taken for granted without justification, which has led, as we shall see, to incorrect extensions of the well-known Lifshitz formula for the Casimir force between two dielectric half-spaces separated by vacuum to the case where the interspace is not empty but also filled with material [11, 23, 24] (see also the textbooks [5, 15, 22] and references therein). Physically, the problems that occur in this context are similar to the ones mentioned above with respect to Eq. (1.1). Irrespective of the calculational details, they may be viewed, for the most frequently considered case of an interspace filled with a (locally responding) dielectric medium, as arising effectively from a formal replacement of the vacuum permittivity with the medium permittivity $\varepsilon_0 \mapsto \varepsilon_0\varepsilon(\mathbf{r},\omega)$ in some energy or stress formula that is valid for free space, which is questionable even if absorption plays a negligible role. The (in general complex-valued) formal action integrals sometimes offered in this context as a supposedly more fundamental starting point (see, e.g., Ref. [25]) are no less questionable. A major aim of this work is to give a fresh approach to the dispersion forces that incorporates from the very beginning a satisfactory description of the macroscopic bodies involved, and, moreover, does not rely on questionable energy or stress expressions for the macroscopic electromagnetic field inside media.

In order to be able to present a theory of dispersion forces with a sound basis and a sufficiently broad range of applicability, we develop, in the first part of the thesis, a very general quantization scheme for the macroscopic electromagnetic field in the presence of linear media, which takes into account not only temporal but also spatial dispersion, as well as absorption. It generalizes previous quantization schemes to a theoretical concept applicable to arbitrary media that respond linearly to the electromagnetic field. The only basic prerequisite is to have available the conductivity tensor of the medium, which enters the macroscopic Maxwell equations as a complex function of frequency, and, in the general case of spatially dispersive media, in a spatially non-local way. We will see that and how previously introduced quantization schemes for diverse classes of media turn out to be limiting cases of our general quantization scheme. Within the framework of this theory we then present, in the second part of the thesis, a unified approach to the calculation of dispersion forces acting on arbitrary ground-state macro- and micro-objects. Since the dispersion forces are, in our opinion, of a purely electromagnetic origin, we regard the (expectation value of the) Lorentz force on appropriately defined charges and currents as the principal quantity from which the dispersion forces should be calculated. More precisely, we consider the ground-state expectation value of the Lorentz force density acting on the charge and current densities attributed to the linearly responding current in linear media, taking fully into account the noise necessarily associated with absorption. From the ground-state Lorentz force density obtained in this way, the dispersion force acting on an arbitrary body or an arbitrary part of it may then be obtained by integration over the respective volume. Our approach thus renders it possible to calculate, unambiguously and in a conceptually clear manner, not only the Casimir force that acts on bodies separated by empty space but also the one which acts on bodies the interspace between which is filled with matter, without any need to resort to debatable energy or stress expressions. Applying the general quantization scheme, we derive very general formulas that enable the calculation of the dispersion force on arbitrary (linearly responding) matter inside a given space region, with arbitrary media present also elsewhere in space. Specializing to bodies that may be viewed as locally responding dielectrics (in the presence of further media), we present a force formula whose applicability ranges from dielectric macro-objects to micro-objects, also including single atoms, without restriction to weakly polarizable material. In particular, this formula enables us to extend the well-known CP-type formula for the force acting on a weakly polarizable (micro-)object to an arbitrary one. It contains, as a special case, the well-known formula for the CP force acting on isolated atoms, and, moreover, it can also be used to calculate the CP force acting on atoms that are constituents of matter, where the neighbouring atoms give rise to a screening effect that diminishes the force. Our theory can also be used to describe—in the very same framework—the vdW force between (ground-state) atoms, in agreement with well-known results. Application of the theory to planar geometries yields extensions of Lifshitz-type formulas and includes also Casimir's well-known original force formula as a limiting case.

Before we begin with the presentation of QED in linearly responding media, we mention that this work is based mainly on the material presented in Refs. [R3],[R6], [R10] and [R11]. In Refs. [R3] and [R6], the theory of dispersion forces has been developed for the case of locally responding magneto-dielectric media. On the basis of the more recent Ref. [R10], we use here the opportunity to generalize a number of results from locally responding magneto-dielectric media to arbitrary linear media with a (possibly) non-local response. For definiteness, we point out here also that the material systems considered in this work are always assumed to be at rest. The electromagnetic dispersion forces have always to be thought of, therefore, as being balanced by other forces. The latter may ultimately result from interactions on length scales that are much smaller than those where the concept of a medium with continuously varying properties is applicable. Hence, despite the fact that the underlying interactions are (first and foremost) again of electromagnetic type, these forces are foreign to macroscopic electrodynamics and not derivable from it. Note that the 'mechanical' forces that might act on a medium on macroscopic scales belong to this category.

Chapter 2

Macroscopic QED in Linearly Responding Media

In both classical and quantum electrodynamics, it is often advisable to divide, at least notionally, the matter that interacts with the electromagnetic field into a part that plays the role of a passive background and a remainder, active part (if any) whose dynamics needs to be followed more explicitly. By means of suitable coarse-graining and averaging procedures, this leads to the well-known framework of Maxwell's macroscopic equations, where the background—the medium—is treated as a continuum, and, quite frequently, by the methods of linear response theory. From this perspective, characterization of the medium is reduced to the prescription of suitable constitutive relations, i.e., appropriate response functions or susceptibilities.

Depending on the specific kinds of media under consideration, it is under many circumstances sufficiently accurate to work with spatially local response functions, taking into account only (temporal) dispersion and absorption in accordance with causality. For conducting and semiconducting media (not to mention plasmas) as well as superconducting materials, however, the spatially local description can be inadequate due to the existence of almost freely movable charge carriers (conduction electrons, excitons, Cooper pairs) in such media. Hence, if one is not willing to restrict one's attention to a crude spatial resolution and/or specific frequency windows, spatial dispersion, i.e., the spatially non-local character of the medium response, generally cannot be disregarded for such media. Electrodynamical problems with the inclusion of spatial dispersion have been considered by various authors in different ways, both on the classical and quantum levels; for classical approaches, see, e.g., Refs. [22, 26–31], for quantum ones see, e.g., Refs. [32, 33].

2.1 Basic Equations

Though it were perfectly legitimate to write quantum-mechanical equations from the outset, it is useful to start the discussions at a classical level. Thus, let us begin with Maxwell's well-known equations for the (frequency-domain) electric field $\underline{\mathbf{E}}(\mathbf{r},\omega)$ and induction field $\underline{\mathbf{B}}(\mathbf{r},\omega)$,

$$\boldsymbol{\nabla}\cdot\underline{\mathbf{B}}(\mathbf{r},\omega)=0, \tag{2.1}$$

$$\boldsymbol{\nabla}\times\underline{\mathbf{E}}(\mathbf{r},\omega)-i\omega\underline{\mathbf{B}}(\mathbf{r},\omega)=0, \tag{2.2}$$

$$\varepsilon_0\boldsymbol{\nabla}\cdot\underline{\mathbf{E}}(\mathbf{r},\omega)=\underline{\rho}(\mathbf{r},\omega), \tag{2.3}$$

$$\mu_0^{-1}\boldsymbol{\nabla}\times\underline{\mathbf{B}}(\mathbf{r},\omega)+i\omega\varepsilon_0\underline{\mathbf{E}}(\mathbf{r},\omega)=\underline{\mathbf{j}}(\mathbf{r},\omega), \tag{2.4}$$

and regard them as classical equations for the time being. For temporal Fourier transforms, we employ the convention

$$\mathbf{E}(\mathbf{r},t)=\int_{-\infty}^{\infty}\mathrm{d}\omega\,e^{-i\omega t}\underline{\mathbf{E}}(\mathbf{r},\omega)=\int_{0}^{\infty}\mathrm{d}\omega\,e^{-i\omega t}\underline{\mathbf{E}}(\mathbf{r},\omega)+\text{c. c.} \tag{2.5}$$

[and accordingly for the other quantities in Eqs. (2.1)–(2.4)], where, as known, the second form makes use of the fact that the field quantities are real-valued [i.e., $\underline{\mathbf{E}}^*(\mathbf{r},\omega)=\underline{\mathbf{E}}(\mathbf{r},-\omega^*)$ etc.]. Equations (2.1)–(2.4) are valid on both microscopic and macroscopic scales and in the presence of arbitrary matter, provided the quantities $\underline{\rho}(\mathbf{r},\omega)$ and $\underline{\mathbf{j}}(\mathbf{r},\omega)$ are identified with the (frequency components of the) total charge and current density, respectively. That is to say, $\underline{\rho}(\mathbf{r},\omega)$ and $\underline{\mathbf{j}}(\mathbf{r},\omega)$ have to account (on a chosen length scale) for all the charges and currents present in space. They are connected by the continuity equation

$$i\omega\underline{\rho}(\mathbf{r},\omega)=\boldsymbol{\nabla}\cdot\underline{\mathbf{j}}(\mathbf{r},\omega), \tag{2.6}$$

the integrability condition between Eqs. (2.3) and (2.4).

If macroscopic amounts of matter are present, it is (usually) not fruitful to interpret Eqs. (2.1)–(2.4) on a microscopic scale; we thus interpret them on a macroscopic scale henceforth. Equations (2.2) and (2.4) may be conveniently combined into a second-order Helmholtz-type equation as

$$\boldsymbol{\nabla}\times\boldsymbol{\nabla}\times\underline{\mathbf{E}}(\mathbf{r},\omega)-\frac{\omega^2}{c^2}\,\underline{\mathbf{E}}(\mathbf{r},\omega)=i\mu_0\omega\underline{\mathbf{j}}(\mathbf{r},\omega). \tag{2.7}$$

Regarding Eq. (2.2) and the continuity equation (2.6) as mere definitions of $\underline{\mathbf{B}}(\mathbf{r},\omega)$ and $\underline{\rho}(\mathbf{r},\omega)$ in terms of $\underline{\mathbf{E}}(\mathbf{r},\omega)$ and $\underline{\mathbf{j}}(\mathbf{r},\omega)$, respectively, it suffices to study Eq. (2.7) in place of the system of Maxwell's equations (2.1)–(2.4). The case of zero frequency (corresponding to strictly static fields) is thereby excluded from consideration, but it can be approached as a limit at later stages if necessary. One might be concerned if important information could be lost in this way. In App. A.1 it is shown that this is not the case—the equations of both electrostatics and magnetostatics can be correctly recovered from the solution to Eq. (2.7) in the static limit $\omega \to 0$. We are thus really permitted to focus on Eq. (2.7), and on the case of non-zero frequencies.

Let us assume that the current density $\underline{\mathbf{j}}(\mathbf{r},\omega)$ can be attributed entirely to the presence of a medium whose internal atomistic structure need not be resolved, with no further (active) sources present in addition to the medium. Assuming that the medium properties are stationary, the frequency components of the total current density may then be described, in the framework of linear response theory, by the constitutive relation

$$\underline{\mathbf{j}}(\mathbf{r},\omega) = \int \mathrm{d}^3 r' \, \boldsymbol{Q}(\mathbf{r},\mathbf{r}',\omega) \cdot \underline{\mathbf{E}}(\mathbf{r}',\omega) + \underline{\mathbf{j}}_{\mathrm{N}}(\mathbf{r},\omega), \tag{2.8}$$

where $\boldsymbol{Q}(\mathbf{r},\mathbf{r}',\omega)$ is the complex, macroscopic conductivity tensor in the frequency domain [31, 34], and $\underline{\mathbf{j}}_{\mathrm{N}}(\mathbf{r},\omega)$ is a Langevin noise source. [Here and throughout, dot products of vectors (boldface) and/or second-rank tensors (boldface and italic) are indicated explicitly; if no intervening symbol is given, the dyadic (or tensor) product is meant.] Equation (2.8) covers all the possible features of a linear medium (in particular, temporal as well as spatial dispersion). According to Onsager's reciprocity theorem [31,34], the conductivity tensor fulfills the reciprocity relation

$$Q_{ij}(\mathbf{r},\mathbf{r}',\omega) = Q_{ji}(\mathbf{r}',\mathbf{r},\omega), \tag{2.9}$$

which we adopt throughout. Except for a translationally invariant (bulk) medium, the spatial arguments $\mathbf{r}$ and $\mathbf{r}'$ of $\boldsymbol{Q}(\mathbf{r},\mathbf{r}',\omega)$ must be kept as two separate variables in general. For chosen ω, $\boldsymbol{Q}(\mathbf{r},\mathbf{r}',\omega)$ is assumed to be the integral kernel of a reasonably well-behaved (integral) operator acting on vector functions in position space. In particular, we assume that $\boldsymbol{Q}(\mathbf{r},\mathbf{r}',\omega)$ tends (sufficiently rapidly) to zero for $|\mathbf{r}-\mathbf{r}'| \to \infty$ and has no strong (i.e., non-integrable) singularities (specifically, for $\mathbf{r}' \to \mathbf{r}$). To allow for the spatially non-dispersive limit, δ-functions and their derivatives must be permitted so that $\boldsymbol{Q}(\mathbf{r},\mathbf{r}',\omega)$ may become a (quasi-)local integral kernel. For the sake of compact notation, superscripts $^{\mathsf{T}}$ and $^{+}$ will be used in the following to indicate transposition and Hermitian conjugation with respect to tensor indices. Since the spatial arguments are not switched by these operations, the operator associated with an integral kernel $\boldsymbol{A}(\mathbf{r},\mathbf{r}')$ is Hermitian (with respect to the usual inner product of vector functions defined in the entire space) if $\boldsymbol{A}(\mathbf{r},\mathbf{r}') = \boldsymbol{A}^{+}(\mathbf{r}',\mathbf{r})$. In particular, an operator associated with a real kernel is Hermitian if it has the reciprocity property $\boldsymbol{A}(\mathbf{r},\mathbf{r}') = \boldsymbol{A}^{\mathsf{T}}(\mathbf{r}',\mathbf{r})$. The decomposition $\boldsymbol{Q}(\mathbf{r},\mathbf{r}',\omega) = \mathrm{Re}\,\boldsymbol{Q}(\mathbf{r},\mathbf{r}',\omega) + i\,\mathrm{Im}\,\boldsymbol{Q}(\mathbf{r},\mathbf{r}',\omega)$ of the conductivity tensor corresponds, due to the reciprocity of $\boldsymbol{Q}(\mathbf{r},\mathbf{r}',\omega)$, to the decomposition of the associated operator into Hermitian and anti-Hermitian parts,

$$\boldsymbol{\sigma}(\mathbf{r},\mathbf{r}',\omega) \equiv \mathrm{Re}\,\boldsymbol{Q}(\mathbf{r},\mathbf{r}',\omega) = \tfrac{1}{2}\big[\boldsymbol{Q}(\mathbf{r},\mathbf{r}',\omega) + \boldsymbol{Q}^{+}(\mathbf{r}',\mathbf{r},\omega)\big], \tag{2.10}$$

$$\boldsymbol{\tau}(\mathbf{r},\mathbf{r}',\omega) \equiv \mathrm{Im}\,\boldsymbol{Q}(\mathbf{r},\mathbf{r}',\omega) = \tfrac{1}{2i}\big[\boldsymbol{Q}(\mathbf{r},\mathbf{r}',\omega) - \boldsymbol{Q}^{+}(\mathbf{r}',\mathbf{r},\omega)\big]. \tag{2.11}$$

Since $\boldsymbol{\sigma}(\mathbf{r},\mathbf{r}',\omega)$ is associated with the dissipation of electromagnetic energy (see, e.g., Refs. [31, 34]), the operator associated with the integral kernel $\boldsymbol{\sigma}(\mathbf{r},\mathbf{r}',\omega)$ is, for real ω, a positive definite operator in the case of absorbing media, i.e.,

$$\int \mathrm{d}^3r \int \mathrm{d}^3r'\, \mathbf{v}^*(\mathbf{r})\cdot\boldsymbol{\sigma}(\mathbf{r},\mathbf{r}',\omega)\cdot\mathbf{v}(\mathbf{r}') > 0 \tag{2.12}$$

for any (quadratically integrable) vector function $\mathbf{v}(\mathbf{r})$. With the exception of Sec. 2.5, we consider absorbing media throughout.

The conductivity tensor $\boldsymbol{Q}(\mathbf{r},\mathbf{r}',\omega)$ is the temporal Fourier transform of a response function,

$$\boldsymbol{Q}(\mathbf{r},\mathbf{r}',\omega) \equiv 2\pi\,\underline{\boldsymbol{Q}}(\mathbf{r},\mathbf{r}',\omega) = \int \mathrm{d}\tau\, e^{i\omega\tau}\,\boldsymbol{Q}(\mathbf{r},\mathbf{r}',\tau), \tag{2.13}$$

where $\boldsymbol{Q}(\mathbf{r},\mathbf{r}',\tau)$ satisfies causality conditions of the type

$$\boldsymbol{Q}(\mathbf{r},\mathbf{r}',\tau) = 0 \quad \text{if} \quad \tau - \cos\eta\,|\mathbf{r}-\mathbf{r}'|/c < 0 \tag{2.14}$$

for chosen $\mathbf{r}$ and $\mathbf{r}'$ and arbitrary directional cosines $\cos\eta$ ($0 \le \cos\eta \le 1$). In particular, for $\cos\eta = 0$, one finds from arguments [34–36] similar to those for the case of spatially locally responding media that, for chosen $\mathbf{r}$ and $\mathbf{r}'$, $\boldsymbol{Q}(\mathbf{r},\mathbf{r}',\omega)$ is analytic in the upper complex ω half-plane, fulfills Kramers–Kronig (Hilbert transform) relations, and satisfies the Schwarz reflection principle

$$\boldsymbol{Q}^*(\mathbf{r},\mathbf{r}',\omega) = \boldsymbol{Q}(\mathbf{r},\mathbf{r}',-\omega^*). \tag{2.15}$$

The (spatio-temporal) conditions obtained by using other values of $\cos\eta$ have been used in Ref. [37] to derive a family of generalized Kramers–Kronig relations (focusing on the case of bulk media); we do not require them here, however. Note that the set of conditions (2.14) is equivalent to the relativistic causality condition [36] that the medium response has to be restricted to the forward light cone (with respect to any chosen Lorentz frame), see Ref. [37].

Inserting Eq.(2.8) in Eq. (2.7), we find that the frequency components of the medium-assisted electric field satisfy the integro-differential equation

$$\begin{aligned}\boldsymbol{\nabla}\times\boldsymbol{\nabla}\times\underline{\mathbf{E}}(\mathbf{r},\omega)-\frac{\omega^2}{c^2}\,\underline{\mathbf{E}}(\mathbf{r},\omega)\\ -\,i\mu_0\omega\int\mathrm{d}^3r'\,\boldsymbol{Q}(\mathbf{r},\mathbf{r}',\omega)\cdot\underline{\mathbf{E}}(\mathbf{r}',\omega)=i\mu_0\omega\underline{\mathbf{j}}_{\mathrm{N}}(\mathbf{r},\omega),\end{aligned}\quad(2.16)$$

which contains as a source term the Langevin noise current density $\underline{\mathbf{j}}_{\mathrm{N}}(\mathbf{r},\omega)$ introduced in Eq. (2.8). A few general remarks on the concept of noise may be in order. From the open-system approach to the description of damped systems, which deals with a system that interacts sufficiently weakly with a sufficiently large number of further (bath) degrees of freedom, it follows quite generally that the net effect of such interaction on the system is to introduce (linear) dissipation, and noise. In this framework, the statistical properties of the noise can be derived from an explicit dynamical model of the overall system. Remarkably, only a few pivotal features of the bath and the interaction are important in this regard, but not the finer details of the explicit model chosen. For a rather broad range of circumstances, it turns out that (classically) the noise can be adequately modeled by a stationary Gaussian stochastic process with zero expectation value. Instead of working with an explicit dynamical model, one may hence directly regard the noise process as given, being defined only through its statistical properties. With this change of perspective, the resulting effective equation of motion for the system is called a Langevin equation—a stochastic equation of motion that features (in general retarded) damping and (in general non-white) noise. In this spirit, we interpret the wave equation (2.16)—or strictly speaking its time-domain counterpart—as an (at this point classical) Langevin equation. As the noise current density $\underline{\mathbf{j}}_{\mathrm{N}}(\mathbf{r},\omega)$ has a vanishing expectation value, by omitting $\underline{\mathbf{j}}_{\mathrm{N}}(\mathbf{r},\omega)$ from Eqs. (2.8) and (2.16) one effectively obtains the corresponding equations for the expectation values, which indicates the connection with ordinary (deterministic) electrodynamics. (Of course, by making a transition to the expectation values in this way, one is discarding the remainder of the probabilistic information as equations involving higher moments are then no longer derivable.) Note that quite generally the demand of consistency of Langevin equations with equilibrium thermodynamics implies quantitative connections between damping and noise known as fluctuation-dissipation relations, see Ref. [34, 38]. For a detailed discussion of Langevin equations—which have been first presented by Langevin in order to give an approach to Brownian motion theory that is "infinitely more simple" (quotation according to Ref. [38]) than the Fokker-Planck equation approach of Einstein and Smoluchowski—we refer the reader to, e.g., Refs. [34, 38–41].

Let us return to Eq. (2.16) and represent its solution in the form

$$\underline{\mathbf{E}}(\mathbf{r},\omega) = i\mu_0\omega \int \mathrm{d}^3r'\, \boldsymbol{G}(\mathbf{r},\mathbf{r}',\omega)\cdot \underline{\mathbf{j}}_{\mathrm{N}}(\mathbf{r}',\omega), \tag{2.17}$$

where $\boldsymbol{G}(\mathbf{r},\mathbf{r}',\omega)$ is the (retarded) Green tensor. It satisfies Eq. (2.16) with the (tensorial) δ-function source,

$$\begin{aligned}\boldsymbol{\nabla}\times\boldsymbol{\nabla}\times \boldsymbol{G}(\mathbf{r},\mathbf{s},\omega) &- \frac{\omega^2}{c^2}\,\boldsymbol{G}(\mathbf{r},\mathbf{s},\omega)\\ &- i\mu_0\omega\int \mathrm{d}^3r'\,\boldsymbol{Q}(\mathbf{r},\mathbf{r}',\omega)\cdot\boldsymbol{G}(\mathbf{r}',\mathbf{s},\omega) = \boldsymbol{I}\delta(\mathbf{r}-\mathbf{s})\end{aligned} \tag{2.18}$$

[$\boldsymbol{I}$, unit tensor], together with the boundary condition at infinity, and has all the attributes of a (Fourier transformed) response function just as $\boldsymbol{Q}(\mathbf{r},\mathbf{r}',\omega)$ has them. In particular, it is analytic in the upper ω half-plane and the Schwarz reflection principle $\boldsymbol{G}^*(\mathbf{r},\mathbf{r}',\omega) = \boldsymbol{G}(\mathbf{r},\mathbf{r}',-\omega^*)$ is valid. Since $\boldsymbol{Q}\,(\mathbf{r},\,\mathbf{r}',\,\omega)$ is reciprocal, so is $\boldsymbol{G}(\mathbf{r},\mathbf{r}',\omega)$, $\boldsymbol{G}(\mathbf{r},\mathbf{r}',\omega) = \boldsymbol{G}^{\mathsf{T}}(\mathbf{r}',\mathbf{r},\omega)$, and, for real ω, the generalized integral relation

$$\mu_0\omega\int \mathrm{d}^3s\int \mathrm{d}^3s'\,\boldsymbol{G}(\mathbf{r},\mathbf{s},\omega)\cdot\boldsymbol{\sigma}(\mathbf{s},\mathbf{s}',\omega)\cdot\boldsymbol{G}^*(\mathbf{s}',\mathbf{r}',\omega) = \mathrm{Im}\,\boldsymbol{G}(\mathbf{r},\mathbf{r}',\omega) \tag{2.19}$$

may be shown to hold (App. A.3). Note that $\boldsymbol{G}(\mathbf{r},\mathbf{r}',\omega)$ is necessarily singular at $\omega = 0$ (for chosen spatial arguments $\mathbf{r}$ and $\mathbf{r}'$). Information about the detailed behavior near $\omega = 0$ of $\boldsymbol{G}(\mathbf{r},\mathbf{r}',\omega)$ [and also of $\boldsymbol{Q}(\mathbf{r},\mathbf{r}',\omega)$], can be obtained from a few basic physical requirements, see App. A.2.

It should be emphasized that Eq. (2.17) is the unique solution to Eq. (2.16)—it is not to be supplemented with any solutions of the source-free version of Eq. (2.16). The source-free version of Eq. (2.16) indeed has no (permissible) solutions existing in the whole space: if the right-hand side of Eq. (2.16) were zero, any (say pulse-like) wave packet observed in some chosen finite space region could only be 'incoming' from spatial infinity, where (because of absorption) it would necessarily have had an infinite amplitude, which is not possible. Clearly, in order to make sure that this argument really works as described, one should in general consider the absorbing medium as filling all of space; free-space regions may then be allowed for at the very end of the calculations. (Strictly speaking, it is already sufficient to imagine the presence of an arbitrarily weakly absorbing medium in all of space outside a finite but arbitrarily large region.) Mathematically, this prescription serves to enforce that the linear integro-differential operator featuring in Eq. (2.16) possesses a unique (both-sided) and bounded inverse operator. To the same end, one may interpret $\boldsymbol{G}(\mathbf{r},\mathbf{r}',\omega)$ as $\boldsymbol{G}(\mathbf{r},\mathbf{r}',\omega+i0)$ wherever necessary.

2.2 Quantization Scheme

Since the solution to Maxwell's equations in the medium specified by Eq. (2.8) is completely embodied in Eq. (2.17), attention may be transferred from the field quantities to the Langevin noise source $\underline{\mathbf{j}}_{\mathrm{N}}(\mathbf{r},\omega)$. Thus, in order to quantize the theory, $\underline{\mathbf{j}}_{\mathrm{N}}(\mathbf{r},\omega)$ is regarded as an operator $[\underline{\mathbf{j}}_{\mathrm{N}}(\mathbf{r},\omega)\mapsto\hat{\underline{\mathbf{j}}}_{\mathrm{N}}(\mathbf{r},\omega)]$, for which commutation relations have to be introduced. The set of operators $\hat{\underline{\mathbf{j}}}_{\mathrm{N}}(\mathbf{r},\omega)$ and $\hat{\underline{\mathbf{j}}}^{\dagger}_{\mathrm{N}}(\mathbf{r},\omega)$ may then be regarded as the dynamical variables of the overall system consisting of the electromagnetic field and the linear medium in terms of which the electromagnetic field operators may be expressed. Clearly, a basic requirement to be met is that the electromagnetic field operators have to obey the correct commutation relations. Since the purpose of the theory is to describe the electromagnetic field interacting with matter, the electromagnetic-field commutation relations to be realized are just the same as those in the underlying microscopic electrodynamics (where they may be viewed as arising from the usual canonical scheme, see, e.g., Ref. [42]), despite the macroscopic viewpoint. Thus, writing the operators of the electric field $\hat{\mathbf{E}}(\mathbf{r})$ and of the magnetic induction field $\hat{\mathbf{B}}(\mathbf{r})$, in a picture-independent manner, as

$$\hat{\mathbf{E}}(\mathbf{r})=\int_0^\infty \mathrm{d}\omega\,\hat{\underline{\mathbf{E}}}(\mathbf{r},\omega)+\mathrm{H.c.},\tag{2.20}$$

$$\hat{\mathbf{B}}(\mathbf{r})=\int_0^\infty \mathrm{d}\omega\,\hat{\underline{\mathbf{B}}}(\mathbf{r},\omega)+\mathrm{H.c.},\tag{2.21}$$

where the respective positive-frequency parts $\underline{\hat{\mathbf{E}}}(\mathbf{r},\omega)$ and $\underline{\hat{\mathbf{B}}}(\mathbf{r},\omega) = (i\omega)^{-1}$ $\boldsymbol{\nabla}\times\underline{\hat{\mathbf{E}}}(\mathbf{r},\omega)$ are expressed in terms of the henceforth operator-valued noise current density as [cf. Eq. (2.17)]

$$\underline{\hat{\mathbf{E}}}(\mathbf{r},\omega) = i\mu_0\omega \int \mathrm{d}^3r' \,\boldsymbol{G}(\mathbf{r},\mathbf{r}',\omega)\cdot \underline{\hat{\mathbf{j}}}_{\mathrm{N}}(\mathbf{r}',\omega), \tag{2.22}$$

$$\underline{\hat{\mathbf{B}}}(\mathbf{r},\omega) = \mu_0 \boldsymbol{\nabla}\times \int \mathrm{d}^3r' \,\boldsymbol{G}(\mathbf{r},\mathbf{r}',\omega)\cdot \underline{\hat{\mathbf{j}}}_{\mathrm{N}}(\mathbf{r}',\omega), \tag{2.23}$$

we have to guarantee the validity of the fundamental equal-time commutator characteristic of the electromagnetic field, i.e.,

$$[\hat{\mathbf{E}}(\mathbf{r}),\hat{\mathbf{B}}(\mathbf{r}')] = i\hbar\boldsymbol{\nabla}\times\boldsymbol{I}\delta(\mathbf{r}-\mathbf{r}')/\varepsilon_0, \tag{2.24}$$

where we have introduced a dyadic notation for commutators. The commutation relation

$$\left[\underline{\hat{\mathbf{j}}}_{\mathrm{N}}(\mathbf{r},\omega),\underline{\hat{\mathbf{j}}}^{\dagger}_{\mathrm{N}}(\mathbf{r}',\omega')\right] = \frac{\hbar\omega}{\pi}\,\delta(\omega-\omega')\,\boldsymbol{\sigma}(\mathbf{r},\mathbf{r}',\omega) \tag{2.25}$$

may be shown to fulfill this requirement, by properly taking into account the properties of $\boldsymbol{G}(\mathbf{r},\mathbf{r}',\omega)$ and $\boldsymbol{Q}(\mathbf{r},\mathbf{r}',\omega)$ [in particular, Eq. (2.19)], along similar lines as known from the spatially local theory (cf. Refs. [43–45]); the derivation is presented in App. A.4. To ensure that the temporal evolution of the electromagnetic field is in accordance with Maxwell's equations, the operators $\underline{\hat{\mathbf{j}}}_{\mathrm{N}}(\mathbf{r},\omega)$ and $\underline{\hat{\mathbf{j}}}^{\dagger}_{\mathrm{N}}(\mathbf{r},\omega)$ have to evolve, in the Heisenberg-picture, like $\sim e^{-i\omega t}$ and $\sim e^{i\omega t}$, respectively. Thus, in order to complete the quantization scheme, a Hamiltonian $\hat{H}$ needs to be introduced [as a functional of $\underline{\hat{\mathbf{j}}}_{\mathrm{N}}(\mathbf{r},\omega)$ and $\underline{\hat{\mathbf{j}}}^{\dagger}_{\mathrm{N}}(\mathbf{r},\omega)$] such that

$$[\underline{\hat{\mathbf{j}}}_{\mathrm{N}}(\mathbf{r},\omega),\hat{H}] = \hbar\omega\,\underline{\hat{\mathbf{j}}}_{\mathrm{N}}(\mathbf{r},\omega), \tag{2.26}$$

which constrains the Hamiltonian to take the form

$$\hat{H} = \pi\int_0^\infty \mathrm{d}\omega\int \mathrm{d}^3r\int \mathrm{d}^3r'\,\underline{\hat{\mathbf{j}}}^{\dagger}_{\mathrm{N}}(\mathbf{r},\omega)\cdot\boldsymbol{\rho}(\mathbf{r},\mathbf{r}',\omega)\cdot\underline{\hat{\mathbf{j}}}_{\mathrm{N}}(\mathbf{r}',\omega), \tag{2.27}$$

apart from an irrelevant c-number contribution. From Eq. (2.26) together with Eqs. (2.25) and (2.27) one can see that $\boldsymbol{\rho}(\mathbf{r},\mathbf{r}',\omega)$ has to be chosen to be the kernel of the integral operator that is the inverse of the integral operator associated with $\boldsymbol{\sigma}(\mathbf{r},\mathbf{r}',\omega)$,

$$\int \mathrm{d}^3s\,\boldsymbol{\rho}(\mathbf{r},\mathbf{s},\omega)\cdot\boldsymbol{\sigma}(\mathbf{s},\mathbf{r}',\omega) = \int \mathrm{d}^3s\,\boldsymbol{\sigma}(\mathbf{r},\mathbf{s},\omega)\cdot\boldsymbol{\rho}(\mathbf{s},\mathbf{r}',\omega) = \boldsymbol{I}\delta(\mathbf{r}-\mathbf{r}'). \tag{2.28}$$

Note that, by means of the correspondence

$$\frac{i}{\varepsilon_0\omega}\,\boldsymbol{Q}(\mathbf{r},\mathbf{r}',\omega) \leftrightarrow \boldsymbol{\chi}(\mathbf{r},\mathbf{r}',\omega), \tag{2.29}$$

where $\boldsymbol{\chi}(\mathbf{r},\mathbf{r}',\omega)$ is the (nonlocal) dielectric susceptibility tensor, the basic commutation relation (2.25) becomes equivalent to the commutation relation derived from a microscopic, linear two-band model of dielectric material [46], which has been used to study the quantized electromagnetic field in spatially dispersive dielectrics [32, 33].

The Hamiltonian (2.27) may clearly be brought to the diagonal form

$$\hat{H} = \int_0^\infty \mathrm{d}\omega\,\hbar\omega \int \mathrm{d}^3r\,\hat{\mathbf{f}}^\dagger(\mathbf{r},\omega)\cdot\hat{\mathbf{f}}(\mathbf{r},\omega) \tag{2.30}$$

known from the spatially local theory, where $\hat{\mathbf{f}}(\mathbf{r},\omega)$ is a bosonic field,

$$\left[\hat{\mathbf{f}}(\mathbf{r},\omega),\hat{\mathbf{f}}^\dagger(\mathbf{r}',\omega')\right] = \delta(\omega-\omega')\boldsymbol{I}\delta(\mathbf{r}-\mathbf{r}'), \tag{2.31}$$

by performing a linear transformation of the variables which we shall assume to be invertible. Writing

$$\underline{\hat{\mathbf{j}}}_{\mathrm{N}}(\mathbf{r},\omega) = \left(\frac{\hbar\omega}{\pi}\right)^{\frac{1}{2}} \int \mathrm{d}^3r'\,\boldsymbol{K}(\mathbf{r},\mathbf{r}',\omega)\cdot\hat{\mathbf{f}}(\mathbf{r}',\omega), \tag{2.32}$$

the diagonalization is achieved and Eqs. (2.25) and (2.31) are rendered equivalent if we choose the integral kernel $\boldsymbol{K}(\mathbf{r},\mathbf{r}',\omega)$ such that, for real ω, the integral equation

$$\int \mathrm{d}^3s\,\boldsymbol{K}(\mathbf{r},\mathbf{s},\omega)\cdot\boldsymbol{K}^+(\mathbf{r}',\mathbf{s},\omega) = \boldsymbol{\sigma}(\mathbf{r},\mathbf{r}',\omega) \tag{2.33}$$

holds, which is guaranteed to possess solutions (see Sec. 2.3) since $\boldsymbol{\sigma}(\mathbf{r},\mathbf{r}',\omega)$ is the integral kernel of a positive definite operator [cf. Eq. (2.12)].

So far we have considered the 'free' medium-assisted electromagnetic field. Its interaction with additional (e.g., atomic) systems can be included in the theory on the basis of the well-known minimal- or multipolar-coupling schemes in the usual way. For instance, let us consider (spinless) point-like, non-relativistic, charged particles (charges Q_a, masses m_a) described in terms of positions $\hat{\mathbf{r}}_a$ and conjugate momenta $\hat{\mathbf{p}}_a$, which interact with the medium-assisted field. One can simply add to the Hamiltonian (2.30) the term

$$\hat{H}' = \sum_a (2m_a)^{-1}[\hat{\mathbf{p}}_a - Q_a\hat{\mathbf{A}}(\hat{\mathbf{r}}_a)]^2 + \hat{W} \tag{2.34}$$

to obtain the total minimal-coupling Hamiltonian $\hat{H}+\hat{H}'$, which [together with commutation relations (2.31) and $[\hat{\mathbf{r}}_a, \hat{\mathbf{p}}_{a'}] = i\hbar\delta_{aa'}\boldsymbol{I}]$ may be used to describe the dynamics of the coupled systems. In Eq. (2.34), $\hat{\mathbf{A}}(\mathbf{r})$ is the vector potential in the Coulomb gauge, i.e., is a purely transverse vector field, $\hat{\mathbf{A}}(\mathbf{r}) = \hat{\mathbf{A}}^{\perp}(\mathbf{r})$, $\hat{\mathbf{A}}^{\|}(\mathbf{r}) = 0$, where the (projection) operations of taking the longitudinal ($\|$) or transverse ($\perp$) parts of a vector field $\mathbf{F}(\mathbf{r})$ are understood, here and below, as

$$\mathbf{F}^{\|}(\mathbf{r}) = \int \mathrm{d}^3r'\, \boldsymbol{\Delta}_{\|}(\mathbf{r}-\mathbf{r}')\cdot\mathbf{F}(\mathbf{r}'), \tag{2.35}$$

$$\mathbf{F}^{\perp}(\mathbf{r}) = \int \mathrm{d}^3r'\, \boldsymbol{\Delta}_{\perp}(\mathbf{r}-\mathbf{r}')\cdot\mathbf{F}(\mathbf{r}'), \tag{2.36}$$

with

$$\boldsymbol{\Delta}_{\|}(\mathbf{r}-\mathbf{r}') = \frac{1}{(2\pi)^3}\int \mathrm{d}^3k\, e^{i\mathbf{k}\cdot(\mathbf{r}-\mathbf{r}')}\,\frac{\mathbf{k}\mathbf{k}}{k^2} \tag{2.37}$$

and

$$\boldsymbol{\Delta}_{\perp}(\mathbf{r}-\mathbf{r}') = \boldsymbol{I}\delta(\mathbf{r}-\mathbf{r}') - \boldsymbol{\Delta}_{\|}(\mathbf{r}-\mathbf{r}') \tag{2.38}$$

being, respectively, the usual longitudinal and transverse (tensor-valued) δ-functions. Writing

$$\hat{\mathbf{A}}(\mathbf{r}) = \int_0^\infty \mathrm{d}\omega\, \underline{\hat{\mathbf{A}}}(\mathbf{r},\omega) + \text{H.c.}, \tag{2.39}$$

we may thus express $\underline{\hat{\mathbf{A}}}(\mathbf{r},\omega)$ in Eq. (2.34) in terms of the transverse part $\underline{\hat{\mathbf{E}}}^{\perp}(\mathbf{r},\omega)$ of $\underline{\hat{\mathbf{E}}}(\mathbf{r},\omega)$ as given by Eq. (2.22),

$$\underline{\hat{\mathbf{A}}}(\mathbf{r},\omega) = (i\omega)^{-1}\underline{\hat{\mathbf{E}}}^{\perp}(\mathbf{r},\omega). \tag{2.40}$$

Taking into account Eq. (2.32), we may eventually express $\hat{\mathbf{A}}(\mathbf{r})$ in terms of the basic bosonic variables $\hat{\mathbf{f}}(\mathbf{r},\omega)$ and $\hat{\mathbf{f}}^{\dagger}(\mathbf{r},\omega)$. The total Coulomb interaction energy $\hat{W}$ in Eq. (2.34) reads

$$\hat{W} = \tfrac{1}{2}\sum_a Q_a\hat{U}(\hat{\mathbf{r}}_a) + \sum_a Q_a\hat{V}(\hat{\mathbf{r}}_a), \tag{2.41}$$

where $\hat{U}(\mathbf{r})$ and $\hat{V}(\mathbf{r})$ are, respectively, the scalar potentials ascribed to the charged particles and the longitudinal part $\hat{\mathbf{E}}^{\|}(\mathbf{r})$ of $\hat{\mathbf{E}}(\mathbf{r})$. Making use of Eq. (2.20) together with Eqs. (2.22) and (2.32), we may express also $\hat{V}(\mathbf{r})$ in terms of $\hat{\mathbf{f}}(\mathbf{r},\omega)$ and $\hat{\mathbf{f}}^{\dagger}(\mathbf{r},\omega)$. For further details, including the multipolar coupling scheme, we refer the reader, e.g., to Ref. [42].

2.3 Natural Variables and Projective Variables

Let us now turn to the problem of constructing the integral kernel $\boldsymbol{K}(\mathbf{r},\mathbf{r}',\omega)$ in Eq. (2.33). For this purpose, we consider the eigenvalue problem

$$\int \mathrm{d}^3r'\,\boldsymbol{\sigma}(\mathbf{r},\mathbf{r}',\omega)\cdot\mathbf{F}(\alpha,\mathbf{r}',\omega)=\sigma(\alpha,\omega)\mathbf{F}(\alpha,\mathbf{r},\omega) \tag{2.42}$$

which, under appropriate regularity assumptions on the conductivity tensor $\boldsymbol{Q}(\mathbf{r},\mathbf{r}',\omega)$ such as those listed below Eq. (2.9), is well-defined. In particular, it features a real (positive) spectrum and a complete set of orthogonal eigensolutions, which we may take to be (δ-)normalized. Note that the real ω plays the role of a parameter here, and α stands for the set of (discrete and/or continuous) quantities needed to label the eigenfunctions. (An α-integration therefore symbolizes multiple summations and/or integrations over all those quantities.) Adopting a continuum notation, we may write

$$\int \mathrm{d}\alpha\,\mathbf{F}(\alpha,\mathbf{r},\omega)\mathbf{F}^*(\alpha,\mathbf{r}',\omega)=\boldsymbol{I}\delta(\mathbf{r}-\mathbf{r}'), \tag{2.43}$$

$$\int \mathrm{d}^3r\,\mathbf{F}^*(\alpha,\mathbf{r},\omega)\cdot\mathbf{F}(\alpha',\mathbf{r},\omega)=\delta(\alpha-\alpha'), \tag{2.44}$$

and the diagonal expansion of $\boldsymbol{\sigma}(\mathbf{r},\mathbf{r}',\omega)$ reads

$$\boldsymbol{\sigma}(\mathbf{r},\mathbf{r}',\omega)=\int \mathrm{d}\alpha\,\sigma(\alpha,\omega)\mathbf{F}(\alpha,\mathbf{r},\omega)\mathbf{F}^*(\alpha,\mathbf{r}',\omega), \tag{2.45}$$

which resembles the expansion of the dielectric susceptibility in the microscopic theory [46] mentioned after Eq. (2.29). Substituting Eq. (2.45) into Eq. (2.33), we may construct an integral kernel $\boldsymbol{K}(\mathbf{r},\mathbf{r}',\omega)$ in the form of

$$\boldsymbol{K}(\mathbf{r},\mathbf{r}',\omega)=\int \mathrm{d}\alpha\,\sigma^{\frac{1}{2}}(\alpha,\omega)\,\mathbf{F}(\alpha,\mathbf{r},\omega)\mathbf{F}^*(\alpha,\mathbf{r}',\omega), \tag{2.46}$$

where we choose $\sigma^{1/2}(\alpha,\omega)>0$ so that the operator associated with $\boldsymbol{K}(\mathbf{r},\mathbf{r}',\omega)$ is the positive, Hermitian square-root of the operator associated with $\boldsymbol{\sigma}\,(\mathbf{r},\mathbf{r}',\omega)$. Obviously, this solution to Eq. (2.33) is not unique, since any other kernel of the form

$$\boldsymbol{K}'(\mathbf{r},\mathbf{r}',\omega)=\int \mathrm{d}^3s\,\boldsymbol{K}(\mathbf{r},\mathbf{s},\omega)\cdot\,\boldsymbol{V}(\mathbf{s},\mathbf{r}',\omega) \tag{2.47}$$

with $\boldsymbol{V}(\mathbf{r},\mathbf{s},\omega)$ satisfying

$$\int \mathrm{d}^3s\ \boldsymbol{V}(\mathbf{r},\mathbf{s},\omega)\cdot \boldsymbol{V}^{+}(\mathbf{r}',\mathbf{s},\omega) = \boldsymbol{I}\delta(\mathbf{r}-\mathbf{r}') \tag{2.48}$$

also obeys Eq. (2.33). As we are interested in invertible transformations (2.32), the operator corresponding to $\boldsymbol{V}(\mathbf{r},\mathbf{s},\omega)$ should be invertible as well, so that we can replace Eq. (2.48) with the stronger unitarity condition

$$\int \mathrm{d}^3s\ \boldsymbol{V}^{+}(\mathbf{s},\mathbf{r},\omega)\cdot \boldsymbol{V}(\mathbf{s},\mathbf{r}',\omega) = \int \mathrm{d}^3s\ \boldsymbol{V}(\mathbf{r},\mathbf{s},\omega)\cdot \boldsymbol{V}^{+}(\mathbf{r}',\mathbf{s},\omega) = \boldsymbol{I}\delta(\mathbf{r}-\mathbf{r}'). \tag{2.49}$$

Obviously, the transition from $\boldsymbol{K}(\mathbf{r},\mathbf{r}',\omega)$ to $\boldsymbol{K}'(\mathbf{r},\mathbf{r}',\omega)$ according to Eq. (2.47) can be viewed alternatively as a redefinition of the dynamical variables $\hat{\mathbf{f}}(\mathbf{r},\omega)$ and $\hat{\mathbf{f}}^{\dagger}(\mathbf{r},\omega)$ according to

$$\hat{\mathbf{f}}(\mathbf{r},\omega) = \int \mathrm{d}^3r'\ \boldsymbol{V}(\mathbf{r},\mathbf{r}',\omega)\cdot \hat{\mathbf{f}}'(\mathbf{r}',\omega), \tag{2.50}$$

$$\hat{\mathbf{f}}^{\dagger}(\mathbf{r},\omega) = \int \mathrm{d}^3r'\ \boldsymbol{V}^{*}(\mathbf{r},\mathbf{r}',\omega)\cdot \hat{\mathbf{f}}'^{\dagger}(\mathbf{r}',\omega). \tag{2.51}$$

Inserting Eq. (2.50) into Eq. (2.32) yields

$$\underline{\hat{\mathbf{j}}}_{\mathrm{N}}(\mathbf{r},\omega) = \left(\frac{\hbar\omega}{\pi}\right)^{\frac{1}{2}} \int \mathrm{d}^3r'\,\boldsymbol{K}'(\mathbf{r},\mathbf{r}',\omega)\cdot \hat{\mathbf{f}}'(\mathbf{r}',\omega), \tag{2.52}$$

where $\boldsymbol{K}'(\mathbf{r},\mathbf{r}',\omega)$ is just given by Eq. (2.47). From this point of view, the significance of replacing Eq. (2.48) with Eq. (2.49) is that the variables $\hat{\mathbf{f}}'(\mathbf{r},\omega)$ and $\hat{\mathbf{f}}'^{\dagger}(\mathbf{r},\omega)$ are uniquely expressible in terms of the $\hat{\mathbf{f}}(\mathbf{r},\omega)$ and $\hat{\mathbf{f}}^{\dagger}(\mathbf{r},\omega)$, and so are on an equal footing with them—the unitary operator associated with $\boldsymbol{V}(\mathbf{r},\mathbf{r}',\omega)$ uniquely maps a set of bosonic variables onto a fully equivalent set of bosonic variables. Hence, $\boldsymbol{V}(\mathbf{r},\mathbf{r}',\omega)$ may be thought of as being included in the chosen set of dynamical variables. Although it is not always advisable to do so (see Sec. 2.4), it is thus in principle sufficient to base the considerations on the Hermitian operator associated with the integral kernel $\boldsymbol{K}(\mathbf{r},\mathbf{r}',\omega)$ as defined by Eq. (2.46). It is worth noting that if the operator associated with $\boldsymbol{K}'(\mathbf{r},\mathbf{r},\omega)$ as defined by Eq. (2.47) happens to be Hermitian, it can differ from the (Hermitian) operator associated with $\boldsymbol{K}(\mathbf{r},\mathbf{r}',\omega)$ only by the trivial type of unitary transformation that merely replaces some of the basis functions $\mathbf{F}(\alpha,\mathbf{r},\omega)$ with $-\mathbf{F}(\alpha,\mathbf{r},\omega)$. Conversely, this shows that any (in this sense) non-trivial $\boldsymbol{V}(\mathbf{r},\mathbf{r}',\omega)$ necessarily yields a non-Hermitian $\boldsymbol{K}'(\mathbf{r},\mathbf{r}',\omega)$ (see Ref. [R10] for a proof of these assertions).

Inserting Eq. (2.46) into Eq. (2.32), we find that

$$\underline{\hat{\mathbf{j}}}_{\mathrm{N}}(\mathbf{r},\omega) = \left(\frac{\hbar\omega}{\pi}\right)^{\frac{1}{2}} \int \mathrm{d}\alpha\, \sigma^{\frac{1}{2}}(\alpha,\omega)\, \mathbf{F}(\alpha,\mathbf{r},\omega)\hat{g}(\alpha,\omega), \tag{2.53}$$

where we have introduced the new variables

$$\hat{g}(\alpha,\omega) = \int \mathrm{d}^3 r\, \mathbf{F}^*(\alpha,\mathbf{r},\omega)\cdot \hat{\mathbf{f}}(\mathbf{r},\omega)\,, \tag{2.54}$$

referred to as the natural variables in the following. Needless to say that they are again of bosonic type,

$$\left[\hat{g}(\alpha,\omega), \hat{g}^\dagger(\alpha',\omega')\right] = \delta(\alpha-\alpha')\delta(\omega-\omega'). \tag{2.55}$$

Since the transformation (2.54) does not mix different ω components, the Hamiltonian (2.30) is still diagonal when expressed in terms of the natural variables,

$$\hat{H} = \int_0^\infty \mathrm{d}\omega\, \hbar\omega \int \mathrm{d}\alpha\, \hat{g}^\dagger(\alpha,\omega)\hat{g}(\alpha,\omega), \tag{2.56}$$

as can be easily seen by inverting Eq. (2.54),

$$\hat{\mathbf{f}}(\mathbf{r},\omega) = \int \mathrm{d}\alpha\, \mathbf{F}(\alpha,\mathbf{r},\omega)\hat{g}(\alpha,\omega), \tag{2.57}$$

and combining with Eq. (2.30), on recalling Eq. (2.44).

Let us organize the set of eigenfunctions $\mathbf{F}(\alpha,\mathbf{r},\omega)$ into (a discrete number of) subsets labeled by λ ($\lambda = 1,2,\ldots,\Lambda$). With the notation $\alpha \mapsto (\lambda,\beta)$, Eq. (2.57) then reads

$$\hat{\mathbf{f}}(\mathbf{r},\omega) = \sum_\lambda \hat{\mathbf{f}}_\lambda(\mathbf{r},\omega), \tag{2.58}$$

where

$$\hat{\mathbf{f}}_\lambda(\mathbf{r},\omega) = \int \mathrm{d}\beta\, \mathbf{F}_\lambda(\beta,\mathbf{r},\omega)\hat{g}_\lambda(\beta,\omega). \tag{2.59}$$

The operators associated with the integral kernels

$$\boldsymbol{P}_\lambda(\mathbf{r},\mathbf{r}',\omega) = \int \mathrm{d}\beta\, \mathbf{F}_\lambda(\beta,\mathbf{r},\omega)\mathbf{F}^*_\lambda(\beta,\mathbf{r}',\omega) \tag{2.60}$$

form a complete set of orthogonal projectors. Obviously, these projectors and the operators associated with $\boldsymbol{\sigma}(\mathbf{r},\mathbf{r}',\omega)$ and $\boldsymbol{K}(\mathbf{r},\mathbf{r}',\omega)$ as given by Eq. (2.46) are commuting quantities. It is not difficult to see that the variables

$$\hat{\mathbf{f}}_\lambda(\mathbf{r},\omega) = \int \mathrm{d}^3 r'\, \boldsymbol{P}_\lambda(\mathbf{r},\mathbf{r}',\omega)\cdot \hat{\mathbf{f}}(\mathbf{r}',\omega) = \int \mathrm{d}\beta\, \mathbf{F}_\lambda(\beta,\mathbf{r},\omega)\hat{g}_\lambda(\beta,\omega), \tag{2.61}$$

referred to as projective variables in the following, obey the non-bosonic commutation relation

$$\left[\hat{\mathbf{f}}_\lambda(\mathbf{r},\omega),\hat{\mathbf{f}}^\dagger_{\lambda'}(\mathbf{r}',\omega')\right] = \delta_{\lambda\lambda'}\delta(\omega-\omega')\boldsymbol{P}_\lambda(\mathbf{r},\mathbf{r}',\omega), \tag{2.62}$$

and the Hamiltonian (2.30) expressed in terms of the projective variables reads as

$$\hat{H} = \sum_\lambda \int_0^\infty \mathrm{d}\omega\,\hbar\omega \int \mathrm{d}^3r\,\hat{\mathbf{f}}^\dagger_\lambda(\mathbf{r},\omega)\cdot\hat{\mathbf{f}}_\lambda(\mathbf{r},\omega). \tag{2.63}$$

From Eqs.(2.62) and (2.63) it then follows that

$$\begin{aligned}\left[\hat{\mathbf{f}}_\lambda(\mathbf{r},\omega),\hat{H}\right] &= \hbar\omega\int \mathrm{d}^3r'\,\boldsymbol{P}_\lambda(\mathbf{r},\mathbf{r}',\omega)\cdot\hat{\mathbf{f}}_\lambda(\mathbf{r}',\omega)\\ &= \hbar\omega\,\hat{\mathbf{f}}_\lambda(\mathbf{r},\omega).\end{aligned} \tag{2.64}$$

Inserting Eq. (2.58) in Eq. (2.32), we obtain

$$\hat{\underline{\mathbf{j}}}_{\mathrm{N}}(\mathbf{r},\omega) = \sum_\lambda \hat{\underline{\mathbf{j}}}_{\mathrm{N}\lambda}(\mathbf{r},\omega), \tag{2.65}$$

where the $\hat{\underline{\mathbf{j}}}_{\mathrm{N}\lambda}(\mathbf{r},\omega)$ are given by

$$\hat{\underline{\mathbf{j}}}_{\mathrm{N}\lambda}(\mathbf{r},\omega) = \left(\frac{\hbar\omega}{\pi}\right)^{\frac{1}{2}} \int \mathrm{d}^3r'\,\boldsymbol{K}_\lambda(\mathbf{r},\mathbf{r}',\omega)\cdot\hat{\mathbf{f}}_\lambda(\mathbf{r}',\omega), \tag{2.66}$$

with

$$\boldsymbol{K}_\lambda(\mathbf{r},\mathbf{r}',\omega) = \int \mathrm{d}^3s\,\boldsymbol{P}_\lambda(\mathbf{r},\mathbf{s},\omega)\cdot\boldsymbol{K}(\mathbf{s},\mathbf{r}',\omega) = \int \mathrm{d}^3s\,\boldsymbol{K}(\mathbf{r},\mathbf{s},\omega)\cdot\boldsymbol{P}_\lambda(\mathbf{s},\mathbf{r}',\omega). \tag{2.67}$$

Recalling Eq. (2.62), we can easily see that

$$\left[\hat{\underline{\mathbf{j}}}_{\mathrm{N}\lambda}(\mathbf{r},\omega),\hat{\underline{\mathbf{j}}}^\dagger_{\mathrm{N}\lambda'}(\mathbf{r}',\omega')\right] = \frac{\hbar\omega}{\pi}\delta_{\lambda\lambda'}\delta(\omega-\omega')\boldsymbol{\sigma}_\lambda(\mathbf{r},\mathbf{r}',\omega), \tag{2.68}$$

where $\boldsymbol{\sigma}_\lambda(\mathbf{r},\mathbf{r}',\omega)$ is defined by the equation obtained from Eq. (2.67) by formally replacing $\boldsymbol{K}_\lambda$ with $\boldsymbol{\sigma}_\lambda$ and $\boldsymbol{K}$ with $\boldsymbol{\sigma}$. Summation of Eq. (2.68) over λ and λ' leads back to Eq. (2.25), so that the two equations are equivalent.

At this stage, we observe that there is the option to base the quantization scheme directly on Eqs. (2.63), (2.65), and (2.66), regarding the variables $\hat{\mathbf{f}}_\lambda(\mathbf{r},\omega)$ and $\hat{\mathbf{f}}^\dagger_\lambda(\mathbf{r},\omega)$ as the basic dynamical variables of the theory and assigning to them bosonic commutation relations

$$\left[\hat{\mathbf{f}}_\lambda(\mathbf{r},\omega),\hat{\mathbf{f}}^\dagger_{\lambda'}(\mathbf{r}',\omega')\right] = \delta_{\lambda\lambda'}\delta(\omega-\omega')\boldsymbol{I}\delta(\mathbf{r}-\mathbf{r}') \tag{2.69}$$

in place of Eq. (2.62). Note that, in so doing, back reference from the variables $\hat{\mathbf{f}}_\lambda(\mathbf{r},\omega)$ to the original variables $\hat{\mathbf{f}}(\mathbf{r},\omega)$ is not possible anymore. As can be seen from Eqs. (2.66) and (2.67), Eq. (2.68) is satisfied also when the $\hat{\mathbf{f}}_\lambda(\mathbf{r},\omega)$ and $\hat{\mathbf{f}}_\lambda^\dagger(\mathbf{r},\omega)$ are considered as bosonic variables, from which it follows [via Eq. (2.65)] that Eq. (2.25) also still holds and, as before, this implies that the correct electromagnetic-field commutation relations hold. The second line of Eq. (2.64) remains of course also true so that the correct time evolution is ensured as well.

Since the state space attributed to the bosonic variables $\hat{\mathbf{f}}_\lambda(\mathbf{r},\omega)$ and $\hat{\mathbf{f}}_\lambda^\dagger(\mathbf{r},\omega)$ is, in general, different from the state space attributed to the original variables $\hat{\mathbf{f}}(\mathbf{r},\omega)$ and $\hat{\mathbf{f}}^\dagger(\mathbf{r},\omega)$ [or, equivalently, attributed to $\hat{g}_\lambda(\beta,\omega)$ and $\hat{g}_\lambda^\dagger(\beta,\omega)$], the allowable states must be restricted, by ruling out certain coherent superpositions of states in the sense of a super-selection rule. In App. A.5, we show that the condition imposed on the states may be described by means of a set of projectors $\hat{P}_\lambda$ such that the allowable states $|\psi\rangle$ can be characterized by

$$\hat{P}_\lambda\,|\psi\rangle = |\psi\rangle \;\;\forall\,\lambda, \tag{2.70}$$

where the action of the projectors $\hat{P}_\lambda$ in state space is closely related to the action of the projectors associated with the kernels (2.60) in position space. As a result, if the total Hamiltonian $\hat{H}_{\mathrm{tot}}$ composed of the Hamiltonian (2.63) and possible interaction terms (in the case where additional, active sources are present) commutes with all of the projectors $\hat{P}_\lambda$,

$$\left[\hat{P}_\lambda, \hat{H}_{\mathrm{tot}}\right] = 0 \;\forall\,\lambda, \tag{2.71}$$

then allowable states remain allowable in the course of time, and the option of treating the $\hat{\mathbf{f}}_\lambda(\mathbf{r},\omega)$ and $\hat{\mathbf{f}}_\lambda^\dagger(\mathbf{r},\omega)$ as bosonic variables can be safely exercised. Clearly, all the observables of interest should then also commute with the $\hat{P}_\lambda$ so that no transition matrix elements between states belonging to different subspaces, i.e., between spaces attributed to different λ values, can ever come into play.

One can also consider decompositions of $\hat{\underline{\mathbf{j}}}_{\mathrm{N}}(\mathbf{r},\omega)$, where in place of the $\hat{\underline{\mathbf{j}}}_{\mathrm{N}\lambda}(\mathbf{r},\omega)$ introduced above other quantities $\hat{\underline{\mathbf{J}}}_{\mathrm{N}\lambda}(\mathbf{r},\omega)$ subject to the condition

$$\sum_\lambda \hat{\underline{\mathbf{J}}}_{\mathrm{N}\lambda}(\mathbf{r},\omega) = \sum_\lambda \hat{\underline{\mathbf{j}}}_{\mathrm{N}\lambda}(\mathbf{r},\omega) \tag{2.72}$$

are introduced, whose commutation relations may be quite different from those of the $\hat{\underline{\mathbf{j}}}_{\mathrm{N}\lambda}(\mathbf{r},\omega)$. Obviously, the total noise current density $\hat{\underline{\mathbf{j}}}_{\mathrm{N}}(\mathbf{r},\omega)$ as given by Eq. (2.65) and the commutation relation (2.25) are not changed by such a transformation, briefly referred to as gauge transformation in the following. Moreover, since, with regard to Eq. (2.25), only the sum of the commutators $[\hat{\underline{\mathbf{J}}}_{\mathrm{N}\lambda}(\mathbf{r},\omega),\hat{\underline{\mathbf{J}}}^{\dagger}_{\mathrm{N}\lambda'}(\mathbf{r}',\omega')]$ over all λ and λ' is relevant, every chosen set of (algebraically consistent) commutators $[\hat{\underline{\mathbf{J}}}_{\mathrm{N}\lambda}(\mathbf{r},\omega),\hat{\underline{\mathbf{J}}}^{\dagger}_{\mathrm{N}\lambda'}(\mathbf{r}',\omega')]$ which leads to Eq. (2.25) yields, in principle, a consistent quantization scheme in its own right. A 'substructure below' Eq. (2.25) can hence be introduced with some arbitrariness, but since the various available alternatives are not necessarily equivalent to each other, a specific one should not be favored in the absence of good (physical) motivation. In contrast, if the observables of interest—including the Hamiltonian—can be viewed as functionals of $\hat{\underline{\mathbf{j}}}_{\mathrm{N}}(\mathbf{r},\omega)$ [rather than of the individual $\hat{\underline{\mathbf{J}}}_{\mathrm{N}\lambda}(\mathbf{r},\omega)$], Eqs. (2.25) and (2.27) can be regarded, in view of the fluctuation-dissipation theorem(s) (see, e.g., Ref. [34]), as being unique, and hence, as invariable fundament of the theory.

From the above, it may be reasonable to widen the notion of projective variables as follows. If, for a chosen (physically motivated) decomposition of the noise current density, it is possible to relate (linearly) the $\hat{\underline{\mathbf{J}}}_{\mathrm{N}\lambda}(\mathbf{r},\omega)$ in Eq. (2.72) to (new) variables $\hat{\mathbf{f}}_{\lambda}(\mathbf{r},\omega)$ such that, upon considering the latter as bosonic variables, the validity of the basic equations (2.25) and (2.26) is ensured, then the specific quantization scheme so obtained may be regarded as arising from the general quantization scheme by excluding certain types of (superposition) states from state space, and restricting the dynamics (as well as the allowable observables) accordingly. The $\hat{\mathbf{f}}_{\lambda}(\mathbf{r},\omega)$ may then be seen as projective variables in a wider sense.

2.4 Different Classes of Media

We proceed to show that rather different classes of media (usually studied separately) fit into the general quantization scheme developed in Sec. 2.2. The main task to be performed is the solution of the eigenvalue problem (2.42), which requires knowledge of $\boldsymbol{\sigma}(\mathbf{r},\mathbf{r}',\omega)$ for the specific medium under consideration. In two limiting cases, the exact solution to Eq. (2.42) can be given straightforwardly, namely, in the case of an inhomogeneous medium without spatial dispersion and in the case of a homogeneous medium that shows spatial dispersion. Let us, therefore, first examine these two cases in detail before considering more general situations.

2.4.1 Spatially Non-Dispersive Inhomogeneous Media

The complete neglect of spatial dispersion means to regard the medium response, i.e., $\boldsymbol{Q}(\mathbf{r},\mathbf{r}',\omega)$, as being strictly local. If this is assumed, we have

$$\boldsymbol{\sigma}(\mathbf{r},\mathbf{r}',\omega)=\boldsymbol{\sigma}(\mathbf{r},\omega)\delta(\mathbf{r}-\mathbf{r}'), \tag{2.73}$$

where $\boldsymbol{\sigma}(\mathbf{r},\omega)$ can be written in diagonal form as

$$\boldsymbol{\sigma}(\mathbf{r},\omega)=\sum_{i=1}^{3}\sigma_i(\mathbf{r},\omega)\,\mathbf{e}_i(\mathbf{r},\omega)\mathbf{e}_i^*(\mathbf{r},\omega), \tag{2.74}$$

with $\mathbf{e}_i(\mathbf{r},\omega)$ $(i=1,2,3)$ being orthogonal unit vectors. Hence, the eigenvalues $\sigma(\alpha,\omega)$ and eigenfunctions $\mathbf{F}(\alpha,\mathbf{r},\omega)$ of the operator associated with $\boldsymbol{\sigma}(\mathbf{r},\mathbf{r}',\omega)$ read $[\alpha\mapsto(i,\mathbf{s})]$ $\sigma_i(\mathbf{s},\omega)$ and

$$\mathbf{F}_i(\mathbf{s},\mathbf{r},\omega)=\mathbf{e}_i(\mathbf{s},\omega)\delta(\mathbf{s}-\mathbf{r}), \tag{2.75}$$

respectively. Equation (2.46) then becomes

$$\boldsymbol{K}(\mathbf{r},\mathbf{r}',\omega)=\boldsymbol{K}(\mathbf{r},\omega)\delta(\mathbf{r}-\mathbf{r}'), \tag{2.76}$$

where

$$\boldsymbol{K}(\mathbf{r},\omega)=\sum_{i=1}^{3}\sigma_i^{1/2}(\mathbf{r},\omega)\,\mathbf{e}_i(\mathbf{r},\omega)\mathbf{e}_i^*(\mathbf{r},\omega), \tag{2.77}$$

and Eq. (2.32) takes the form

$$\underline{\hat{\mathbf{j}}}_{\mathrm{N}}(\mathbf{r},\omega)=\left(\frac{\hbar\omega}{\pi}\right)^{\frac{1}{2}}\boldsymbol{K}(\mathbf{r},\omega)\cdot\hat{\mathbf{f}}(\mathbf{r},\omega), \tag{2.78}$$

which just yields the well-known quantization scheme for a locally responding, possibly anisotropic dielectric material [43, 44], upon identifying $\boldsymbol{\sigma}(\mathbf{r},\omega)$ $=\varepsilon_0\omega\,\mathrm{Im}\,\boldsymbol{\chi}(\mathbf{r},\omega)$, with $\boldsymbol{\chi}(\mathbf{r},\omega)$ being the (local) dielectric susceptibility tensor [cf. Eq. (2.29)]. The natural variables $\hat{g}_i(\mathbf{r},\omega)$ are here simply the components of $\hat{\mathbf{f}}(\mathbf{r},\omega)$ along the principal axes of the medium, which may in general vary with position and frequency,

$$\hat{g}_i(\mathbf{r},\omega)=\mathbf{e}_i^*(\mathbf{r},\omega)\cdot\hat{\mathbf{f}}(\mathbf{r},\omega), \tag{2.79}$$

$$\hat{\mathbf{f}}(\mathbf{r},\omega)=\sum_{i=1}^{3}\mathbf{e}_i(\mathbf{r},\omega)\hat{g}_i(\mathbf{r},\omega). \tag{2.80}$$

It is worth noting that the concept of principal axes of an absorbing medium is ambiguous in general, since two sets of principal axes—defined by either the orthogonal eigenvectors of the real part or those of the imaginary part of the (local) conductivity tensor—can be considered. Conventionally, the principal axes are discussed only for (spatially non-dispersive) anisotropic dielectrics with negligible absorption (see, e.g., Ref. [47]). Recalling Eq. (2.29), we can conclude that the 'conventional' set of orthogonal axes is the one obtained from the eigenvectors of the imaginary part of the (local) conductivity tensor, whereas the $\mathbf{e}_i(\mathbf{r},\omega)$ introduced in Eq. (2.74) refer to the real part. The two sets of principal axes cannot in general be chosen to agree, but they can in the particular case where their dependence on frequency (called dispersion of the axes) may be ignored, as may be shown using the Kramers-Kronig relations. According to Ref. [47], dispersion of the axes can exist in a (crystalline) medium only if its symmetry, on microscopic scales, does not determine a preferential orthogonal triplet of directions.

Identifying the index λ introduced in Eq. (2.58) with i and assuming that $\sigma_i(\mathbf{r},\omega)\neq\sigma_{i'}(\mathbf{r},\omega)$ for $i\neq i'$, one can define, according to Eq. (2.60), the three projection kernels

$$\boldsymbol{P}_i(\mathbf{r},\mathbf{r}',\omega)=\mathbf{e}_i(\mathbf{r},\omega)\mathbf{e}_i^*(\mathbf{r}',\omega)\delta(\mathbf{r}-\mathbf{r}') \tag{2.81}$$

which, according to Eq. (2.61), give rise to three sets of projective variables,

$$\hat{\mathbf{f}}_i(\mathbf{r},\omega)=\mathbf{e}_i(\mathbf{r},\omega)\hat{g}_i(\mathbf{r},\omega). \tag{2.82}$$

As long as the projective variables are not coupled to each other—which is obviously the case for the 'free' system governed by the Hamiltonian (2.64)—they can be regarded as being of bosonic type. In this case, instead of using the original set of bosonic variables $\hat{\mathbf{f}}(\mathbf{r},\omega)$ and $\hat{\mathbf{f}}^\dagger(\mathbf{r},\omega)$, one can use three sets of bosonic variables $\hat{\mathbf{f}}_i(\mathbf{r},\omega)$ and $\hat{\mathbf{f}}_i^\dagger(\mathbf{r},\omega)$ associated with the three principal axes of the dielectric medium at each space point.

If two of the three eigenvalues $\sigma_i(\mathbf{r},\omega)$ coincide (uniaxial medium), the two corresponding projection kernels $\boldsymbol{P}_i(\mathbf{r},\mathbf{r}',\omega)$ should be combined into one projector (projecting on the plane perpendicular to the distinguished axis of the medium), thereby reducing the number of sets of projective variables to two. Clearly, if the three eigenvalues $\sigma_i(\mathbf{r},\omega)$ all coincide (isotropic medium), the three projection kernels $\boldsymbol{P}_i(\mathbf{r},\mathbf{r}',\omega)$ should be combined to give the unit kernel $\boldsymbol{I}\delta(\mathbf{r}-\mathbf{r}')$, corresponding to the use of the original variables.

2.4.2 Spatially Dispersive Homogeneous Media

In the limiting case of an (infinitely extended) homogeneous medium, $\boldsymbol{Q}$ $(\mathbf{r}, \mathbf{r}', \omega)$ is translationally invariant, i.e., it is a function of the difference $\mathbf{r}-\mathbf{r}'$, and so is then $\boldsymbol{\sigma}(\mathbf{r}, \mathbf{r}', \omega)$. We may therefore represent it as the spatial Fourier transform

$$\boldsymbol{\sigma}(\mathbf{r}, \mathbf{r}', \omega) = \frac{1}{(2\pi)^3} \int \mathrm{d}^3k\, \boldsymbol{\sigma}(\mathbf{k}, \omega) e^{i\mathbf{k}\cdot(\mathbf{r}-\mathbf{r}')}, \tag{2.83}$$

where

$$\boldsymbol{\sigma}(\mathbf{k}, \omega) = \sum_{i=1}^{3} \sigma_i(\mathbf{k}, \omega)\, \mathbf{e}_i(\mathbf{k}, \omega) \mathbf{e}_i^*(\mathbf{k}, \omega), \tag{2.84}$$

with $\mathbf{e}_i(\mathbf{k}, \omega)$ $(i = 1, 2, 3)$ being orthogonal unit vectors. Consequently, the eigenvalues $\sigma(\alpha, \omega)$ and eigenfunctions $\mathbf{F}(\alpha, \mathbf{r}, \omega)$ of the operator associated with $\boldsymbol{\sigma}(\mathbf{r}, \mathbf{r}', \omega)$ are $[\alpha \mapsto (i, \mathbf{k})]$ $\sigma_i(\mathbf{k}, \omega)$ and

$$\mathbf{F}_i(\mathbf{k}, \mathbf{r}, \omega) = (2\pi)^{-3/2} e^{i\mathbf{k}\cdot\mathbf{r}} \mathbf{e}_i(\mathbf{k}, \omega), \tag{2.85}$$

respectively, and Eq. (2.46) reads

$$\boldsymbol{K}(\mathbf{r}, \mathbf{r}', \omega) = \frac{1}{(2\pi)^3} \int \mathrm{d}^3k\, \boldsymbol{K}(\mathbf{k}, \omega) e^{i\mathbf{k}\cdot(\mathbf{r}-\mathbf{r}')}, \tag{2.86}$$

where

$$\boldsymbol{K}(\mathbf{k}, \omega) = \sum_{i=1}^{3} \sigma_i^{1/2}(\mathbf{k}, \omega)\, \mathbf{e}_i(\mathbf{k}, \omega) \mathbf{e}_i^*(\mathbf{k}, \omega). \tag{2.87}$$

Combination of Eqs. (2.32), (2.86), and (2.87) then yields

$$\underline{\mathbf{j}}_{\mathrm{N}}(\mathbf{r}, \omega) = \left(\frac{\hbar\omega}{\pi}\right)^{\frac{1}{2}} \frac{1}{(2\pi)^{3/2}} \sum_{i=1}^{3} \int \mathrm{d}^3k\, e^{i\mathbf{k}\cdot\mathbf{r}} \sigma_i^{1/2}(\mathbf{k}, \omega) \mathbf{e}_i(\mathbf{k}, \omega) \hat{g}_i(\mathbf{k}, \omega), \tag{2.88}$$

where the natural variables $\hat{g}_i(\mathbf{k}, \omega)$ are related to the spatial Fourier components of $\hat{\mathbf{f}}(\mathbf{r}, \omega)$ as

$$\hat{g}_i(\mathbf{k}, \omega) = \frac{1}{(2\pi)^{3/2}} \int \mathrm{d}^3r\, e^{-i\mathbf{k}\cdot\mathbf{r}}\, \mathbf{e}_i^*(\mathbf{k}, \omega) \cdot \hat{\mathbf{f}}(\mathbf{r}, \omega). \tag{2.89}$$

On the basis of the three unit vectors $\mathbf{e}_i(\mathbf{k}, \omega)$, three (different) projection kernels can be introduced,

$$\boldsymbol{P}_i(\mathbf{r}, \mathbf{r}', \omega) = \frac{1}{(2\pi)^3} \int \mathrm{d}^3k\, \mathbf{e}_i(\mathbf{k}, \omega) \mathbf{e}_i^*(\mathbf{k}, \omega)\, e^{i\mathbf{k}\cdot(\mathbf{r}-\mathbf{r}')}, \tag{2.90}$$

provided that $\sigma_i(\mathbf{k},\omega)\neq\sigma_{i'}(\mathbf{k},\omega)$ for $i\neq i'$.

Let us consider the particular case of isotropic media without optical activity in more detail. In this situation, the diagonal form of the tensor $\boldsymbol{\sigma}(\mathbf{k},\omega)$ reads (see Ref. [31])

$$\boldsymbol{\sigma}(\mathbf{k},\omega) = \sigma_{\|}(k,\omega)\frac{\mathbf{k}\mathbf{k}}{k^2} + \sigma_{\perp}(k,\omega)\left(\boldsymbol{I} - \frac{\mathbf{k}\mathbf{k}}{k^2}\right), \tag{2.91}$$

i.e., $\sigma_1(k,\omega)=\sigma_{\|}(k,\omega)$ and $\sigma_2(k,\omega)=\sigma_3(k,\omega)=\sigma_{\perp}(k,\omega)\neq\sigma_{\|}(k,\omega)$, which implies that $\boldsymbol{K}(\mathbf{k},\omega)$, Eq. (2.87), takes the form

$$\boldsymbol{K}(\mathbf{k},\omega) = \sigma_{\|}^{1/2}(k,\omega)\frac{\mathbf{k}\mathbf{k}}{k^2} + \sigma_{\perp}^{1/2}(k,\omega)\left(\boldsymbol{I} - \frac{\mathbf{k}\mathbf{k}}{k^2}\right). \tag{2.92}$$

Thus, the longitudinal and transverse tensorial δ-functions given in Eqs. (2.37) and (2.38) respectively, can be taken as projection kernels,

$$\boldsymbol{P}_{\|(\perp)}(\mathbf{r},\mathbf{r}',\omega) = \boldsymbol{\Delta}_{\|(\perp)}(\mathbf{r}-\mathbf{r}'), \tag{2.93}$$

which may be used to introduce, according to Eq. (2.61), the projective variables

$$\hat{\mathbf{f}}_{\|(\perp)}(\mathbf{r},\omega) = \int \mathrm{d}^3 s\, \boldsymbol{\Delta}_{\|(\perp)}(\mathbf{r}-\mathbf{s})\cdot\hat{\mathbf{f}}(\mathbf{s},\omega). \tag{2.94}$$

Unitarily Equivalent Formulation

As already pointed out in Sec. 2.3, the integral kernel $\boldsymbol{K}(\mathbf{r},\mathbf{r}',\omega)$ in Eq. (2.32) is not uniquely determined by Eq. (2.33), since any other kernel $\boldsymbol{K}'(\mathbf{r},\mathbf{r}',\omega)$ of the form (2.47) [together with Eq. (2.49)] is also an allowed kernel. To illustrate this for the isotropic medium under study, we first note that Eq. (2.91) may be equivalently rewritten as

$$\boldsymbol{\sigma}(\mathbf{k},\omega) = \sigma_{\|}(k,\omega)\boldsymbol{I} - \mathbf{k}\times\gamma(k,\omega)\boldsymbol{I}\times\mathbf{k}, \tag{2.95}$$

where

$$\gamma(k,\omega) = [\sigma_{\perp}(k,\omega) - \sigma_{\|}(k,\omega)]/k^2. \tag{2.96}$$

Since (for real ω) $\sigma_{\|}(k,\omega)$ and $\sigma_{\perp}(k,\omega)$ are both real and positive [in accordance with the requirement that $\boldsymbol{\sigma}(\mathbf{r},\mathbf{r}',\omega)$ be the integral kernel of a positive definite operator], $\gamma(k,\omega)$ is real but its sign is not determined by this requirement. However, if $\gamma(k,\omega)$ is required here and below to be positive throughout, then

$$\boldsymbol{K}'(\mathbf{k},\omega) = \sigma_{\|}^{1/2}(k,\omega)\boldsymbol{I} \pm \gamma^{1/2}(k,\omega)\mathbf{k}\times\boldsymbol{I} \tag{2.97}$$

obeys the equation

$$\boldsymbol{K}'(\mathbf{k},\omega)\cdot\boldsymbol{K}'^{+}(\mathbf{k},\omega)=\boldsymbol{\sigma}(\mathbf{k},\omega). \tag{2.98}$$

Moreover, it can be shown that $\boldsymbol{K}'(\mathbf{k},\omega)$ can be represented in the form

$$\boldsymbol{K}'(\mathbf{k},\omega)=\boldsymbol{K}(\mathbf{k},\omega)\cdot\boldsymbol{V}(\mathbf{k},\omega), \tag{2.99}$$

with

$$\boldsymbol{V}(\mathbf{k},\omega)=\frac{\mathbf{k}\mathbf{k}}{k^2}+\sigma_{\|}^{1/2}(k,\omega)\sigma_{\perp}^{-1/2}(k,\omega)\left(\boldsymbol{I}-\frac{\mathbf{k}\mathbf{k}}{k^2}\right)\pm\gamma^{1/2}(k,\omega)\sigma_{\perp}^{-1/2}(k,\omega)\mathbf{k}\times\boldsymbol{I} \tag{2.100}$$

[$\boldsymbol{V}^{-1}(\mathbf{k},\omega)=\boldsymbol{V}^{+}(\mathbf{k},\omega)$]. Hence, $\boldsymbol{K}'(\mathbf{k},\omega)$ also yields, according to Eq. (2.86), a valid integral kernel $\boldsymbol{K}'(\mathbf{r},\mathbf{r}',\omega)$,

$$\boldsymbol{K}'(\mathbf{r},\mathbf{r}',\omega)=\frac{1}{(2\pi)^3}\int \mathrm{d}^3k\,\boldsymbol{K}'(\mathbf{k},\omega)e^{i\mathbf{k}\cdot(\mathbf{r}-\mathbf{r}')}, \tag{2.101}$$

which is related to the integral kernel $\boldsymbol{K}(\mathbf{r},\mathbf{r}',\omega)$ according to Eq. (2.47), where

$$\boldsymbol{V}(\mathbf{r},\mathbf{r}',\omega)=\frac{1}{(2\pi)^3}\int \mathrm{d}^3k\;\boldsymbol{V}(\mathbf{k},\omega)e^{i\mathbf{k}\cdot(\mathbf{r}-\mathbf{r}')}, \tag{2.102}$$

with the associated operator being unitary. We thus see that the two formulations of the theory based on $\boldsymbol{K}(\mathbf{r},\mathbf{r}',\omega)$ and $\boldsymbol{K}'(\mathbf{r},\mathbf{r}',\omega)$, respectively, are unitarily equivalent. Note that $\boldsymbol{K}'(\mathbf{k},\omega)\neq\boldsymbol{K}'^{+}(\mathbf{k},\omega)$, so that the operator associated with the integral kernel $\boldsymbol{K}'(\mathbf{r},\mathbf{r}',\omega)$ is non-Hermitian. Since the operators associated with $\boldsymbol{K}'(\mathbf{r},\mathbf{r}',\omega)$ [as well as $\boldsymbol{V}(\mathbf{r},\mathbf{r}',\omega)$] and $\boldsymbol{P}_{\|(\perp)}(\mathbf{r},\mathbf{r}',\omega)$ commute, the same projectors may be employed in the two formulations of the theory to introduce projective variables according to Eq. (2.94).

Local Limit: Magnetodielectric Media

Now let us suppose that $\sigma_{\|}(k,\omega)$ and $\gamma(k,\omega)$ in Eq. (2.95) are sufficiently slowly varying functions of k, with well-defined and unique long-wavelength limits $\lim_{k\to0}\sigma_{\|}(k,\omega)=\sigma_{\|}(\omega)>0$ and $\lim_{k\to0}\gamma(k,\omega)=\gamma(\omega)>0$, so that they may be approximated by these limits under the integral in Eq. (2.83) to obtain

$$\boldsymbol{\sigma}(\mathbf{r},\mathbf{r}',\omega)=\sigma_{\|}(\omega)\boldsymbol{I}\delta(\mathbf{r}-\mathbf{r}')-\gamma(\omega)\boldsymbol{\nabla}\times[\boldsymbol{I}\delta(\mathbf{r}-\mathbf{r}')]\times\overleftarrow{\boldsymbol{\nabla}}'. \tag{2.103}$$

It should be pointed out that in the limiting case given by Eq. (2.103) the positive definiteness of the operator associated with $\boldsymbol{\sigma}(\mathbf{r},\mathbf{r}',\omega)$ already implies that $\gamma(\omega)$ must be positive, $\gamma(\omega)>0$; in the general case as given by Eq. (2.83) together with Eqs. (2.95) and (2.96), the positive definiteness of $\boldsymbol{\sigma}(\mathbf{r},\mathbf{r}',\omega)$ does not automatically restrict $\gamma(k,\omega)$ to positive values.

In order to see to what type of medium this $\boldsymbol{\sigma}(\mathbf{r},\mathbf{r}',\omega)$ corresponds, we have to find from Eq. (2.103) the full conductivity tensor $\boldsymbol{Q}(\mathbf{r},\mathbf{r}',\omega)$, which is uniquely possible since Eqs. (2.10) and (2.11) are Hilbert transforms of each other (cf. Sec. 2.2). The full conductivity tensor corresponding to Eq. (2.103) is thus of the form

$$\boldsymbol{Q}(\mathbf{r},\mathbf{r}',\omega) = Q^{(1)}(\omega)\boldsymbol{I}\delta(\mathbf{r}-\mathbf{r}') - Q^{(2)}(\omega)\boldsymbol{\nabla}\times[\boldsymbol{I}\delta(\mathbf{r}-\mathbf{r}')]\times\overleftarrow{\boldsymbol{\nabla}}', \tag{2.104}$$

where $Q^{(1)}(\omega)$ and $Q^{(2)}(\omega)$ are (Fourier-transformed) response functions, both of which are determined by their respective real parts $\sigma_{\|}(\omega)$ and $\gamma(\omega)$. Inserting Eq. (2.104) into Eq. (2.8) and comparing with

$$\hat{\underline{\mathbf{j}}}(\mathbf{r},\omega) = -i\varepsilon_0\omega\,[\varepsilon(\omega)-1]\hat{\underline{\mathbf{E}}}(\mathbf{r},\omega) + \kappa_0\boldsymbol{\nabla}\times\{[1-\kappa(\omega)]\hat{\underline{\mathbf{B}}}(\mathbf{r},\omega)\} + \hat{\underline{\mathbf{j}}}_{\mathrm{N}}(\mathbf{r},\omega) \tag{2.105}$$

$[\hat{\underline{\mathbf{B}}}(\mathbf{r},\omega) = (i\omega)^{-1}\boldsymbol{\nabla}\times\hat{\underline{\mathbf{E}}}(\mathbf{r},\omega)]$, which is the well-known description of a locally responding (homogeneous) magnetodielectric medium, we can make the identifications

$$Q^{(1)}(\omega) = -i\varepsilon_0\omega\,[\varepsilon(\omega)-1] \tag{2.106}$$

and

$$Q^{(2)}(\omega) = -i\kappa_0[1-\kappa(\omega)]/\omega, \tag{2.107}$$

where $\varepsilon(\omega)$ is the permittivity and $\mu(\omega)=\kappa^{-1}(\omega)$ the (paramagnetic) permeability of the medium ($\mu_0=\kappa_0^{-1}$). For real ω, we thus obtain

$$\sigma_{\|}(\omega) = \varepsilon_0\omega\,\mathrm{Im}\,\varepsilon(\omega) \tag{2.108}$$

and

$$\gamma(\omega) = -\kappa_0\mathrm{Im}\,\kappa(\omega)/\omega. \tag{2.109}$$

Note that, because of $\gamma(\omega)>0$, from Eq. (2.109) it follows that $\mathrm{Im}\,\kappa(\omega)<0$ for $\omega>0$, from which it can be shown that $\mu(\omega\to 0)>1$.

At first glance, one might believe (erroneously) that not only paramagnetic $[\mu(\omega\to 0)>1]$ but also diamagnetic $[\mu(\omega\to 0)<1]$ features of a medium (or the combined effect of both) can be consistently described by means of the magnetic permeability $\mu(\omega)$ which is included, as seen above, in the basic linear-response constitutive relation (2.8). However, since diamagnetism is basically a nonlinear effect (as the underlying microscopic Hamiltonian is quadratic in the magnetic induction field), it is beyond the scope of linear response theory. If it is desired to include diamagnetic media in the framework of linear electrodynamics nevertheless, one can regard the magnetic field on which the diamagnetic susceptibility depends as being (the mean value of) an externally controlled field independent of the dynamical variables. Note that the Onsager reciprocity theorem needs to be stated in its generalized form in this case, see Refs. [31, 34]. For a more satisfactory account of diamagnetic media, one should, however, resort to a non-linear response formalism, or to a more microscopic theory.

An obvious solution to Eq. (2.33) with $\boldsymbol{\sigma}(\mathbf{r},\mathbf{r}',\omega)$ given by Eq. (2.103) is provided by

$$\boldsymbol{K}'(\mathbf{r},\mathbf{r}',\omega) = \sigma_{\|}^{1/2}(\omega)\boldsymbol{I}\delta(\mathbf{r}-\mathbf{r}') \mp i\gamma^{1/2}(\omega)\boldsymbol{\nabla}\times\boldsymbol{I}\delta(\mathbf{r}-\mathbf{r}'), \tag{2.110}$$

which corresponds to the kernel (2.101) [together with Eq. (2.97)] when, for an isotropic medium, $\sigma_{\|}(k,\omega)$ and $\gamma(k,\omega)$ are approximated by $\sigma_{\|}(\omega)$ and $\gamma(\omega)$, respectively. The kernel (2.101) [together with Eq. (2.97)] fits well here since it depends in a particularly simple way on those quantities that we have assumed to approach well-defined limits in the derivation that led to Eq. (2.103), a property which can be attributed to the responsible transformation (2.102) [together with (2.100)]. In contrast, the kernel obtained directly from Eq. (2.86) [together with Eq. (2.92)], by first eliminating $\sigma_{\perp}(k,\omega)$ by means of Eq. (2.96) and then approximating $\sigma_{\|}(k,\omega)\mapsto\sigma_{\|}(\omega)$ and $\gamma(k,\omega)\mapsto\gamma(\omega)$, is not unitarily equivalent to Eq. (2.110), as it does not lead to Eq. (2.103) when inserted in Eq. (2.33); it corresponds to a different medium.

Substituting for $\boldsymbol{K}(\mathbf{r},\mathbf{r}',\omega)$ in Eq. (2.32) $\boldsymbol{K}'(\mathbf{r},\mathbf{r}',\omega)$ as given by Eq. (2.110) [together with Eqs. (2.108) and (2.109)], we may explicitly express the noise current density in terms of the bosonic dynamical variables to obtain

$$\hat{\underline{\mathbf{j}}}_{\mathrm{N}}(\mathbf{r},\omega) = \left(\frac{\hbar\varepsilon_0}{\pi}\right)^{\frac{1}{2}}\sqrt{\omega^2\mathrm{Im}\,\varepsilon(\omega)}\,\hat{\mathbf{f}}(\mathbf{r},\omega) \mp i\left(\frac{\hbar\kappa_0}{\pi}\right)^{\frac{1}{2}}\boldsymbol{\nabla}\times\left[\sqrt{-\mathrm{Im}\,\kappa(\omega)}\,\hat{\mathbf{f}}(\mathbf{r},\omega)\right]. \tag{2.111}$$

Since the operators associated with the projection kernels (2.93) commute with the operators associated with Eqs. (2.110) and (2.103), one may introduce the projective variables $\hat{\mathbf{f}}_{\|(\perp)}(\mathbf{r},\omega)$ defined by Eq. (2.94), which corresponds to a decomposition of $\hat{\underline{\mathbf{j}}}_{\mathrm{N}}(\mathbf{r},\omega)$ into longitudinal and transverse parts,

$$\hat{\underline{\mathbf{j}}}_{\mathrm{N}}(\mathbf{r},\omega) = \hat{\underline{\mathbf{j}}}_{\mathrm{N}\|}(\mathbf{r},\omega) + \hat{\underline{\mathbf{j}}}_{\mathrm{N}\perp}(\mathbf{r},\omega), \tag{2.112}$$

where

$$\underline{\hat{\mathbf{j}}}_{\mathrm{N}\parallel}(\mathbf{r},\omega) = \left(\frac{\hbar\varepsilon_0}{\pi}\right)^{\frac{1}{2}} \sqrt{\omega^2 \mathrm{Im}\,\varepsilon(\omega)}\,\hat{\mathbf{f}}_{\parallel}(\mathbf{r},\omega), \tag{2.113}$$

$$\underline{\hat{\mathbf{j}}}_{\mathrm{N}\perp}(\mathbf{r},\omega) = \left(\frac{\hbar\varepsilon_0}{\pi}\right)^{\frac{1}{2}} \sqrt{\omega^2 \mathrm{Im}\,\varepsilon(\omega)}\,\hat{\mathbf{f}}_{\perp}(\mathbf{r},\omega) \\ \mp i \left(\frac{\hbar\kappa_0}{\pi}\right)^{\frac{1}{2}} \boldsymbol{\nabla}\times\left[\sqrt{-\mathrm{Im}\,\kappa(\omega)}\,\hat{\mathbf{f}}_{\perp}(\mathbf{r},\omega)\right]. \tag{2.114}$$

Making use of Eq. (2.61) and identifying therein the projection kernels $\boldsymbol{P}_\lambda(\mathbf{r},\mathbf{r}',\omega)$ with $\boldsymbol{\Delta}_{\parallel(\perp)}(\mathbf{r}-\mathbf{r}')$, one may then proceed as described in Sec. 2.3 and regard the projective variables $\hat{\mathbf{f}}_{\parallel}(\mathbf{r},\omega)$ and $\hat{\mathbf{f}}_{\perp}(\mathbf{r},\omega)$ as being two independent sets of bosonic variables.

Let us briefly make contact with the quantization scheme described in Refs. [43, 45], where $\underline{\hat{\mathbf{j}}}_{\mathrm{N}}(\mathbf{r},\omega)$ is decomposed according to

$$\underline{\hat{\mathbf{j}}}_{\mathrm{N}}(\mathbf{r},\omega) = \underline{\hat{\mathbf{J}}}_{\mathrm{N}e}(\mathbf{r},\omega) + \underline{\hat{\mathbf{J}}}_{\mathrm{N}m}(\mathbf{r},\omega), \tag{2.115}$$

with

$$\underline{\hat{\mathbf{J}}}_{\mathrm{N}e}(\mathbf{r},\omega) = \left(\frac{\hbar\varepsilon_0}{\pi}\right)^{\frac{1}{2}} \sqrt{\omega^2 \mathrm{Im}\,\varepsilon(\omega)}\,\hat{\mathbf{f}}_{e}(\mathbf{r},\omega), \tag{2.116}$$

$$\underline{\hat{\mathbf{J}}}_{\mathrm{N}m}(\mathbf{r},\omega) = \mp i \left(\frac{\hbar\kappa_0}{\pi}\right)^{\frac{1}{2}} \boldsymbol{\nabla}\times\left[\sqrt{-\mathrm{Im}\,\kappa(\omega)}\,\hat{\mathbf{f}}_{m}(\mathbf{r},\omega)\right]. \tag{2.117}$$

The connection between Eqs. (2.112)–(2.114) and Eqs. (2.115)–(2.117) is given by a gauge transformation (cf. Sec. 2.3), which effectively redistributes the first term of Eq. (2.114). It is not difficult to prove that the total noise current density as given by Eq. (2.115) satisfies the correct commutation relation (2.25) [with $\boldsymbol{\sigma}(\mathbf{r},\mathbf{r}',\omega)$ from Eq. (2.103) together with Eqs. (2.108) and (2.109)] if $\hat{\mathbf{f}}_e(\mathbf{r},\omega)$ and $\hat{\mathbf{f}}_m(\mathbf{r},\omega)$ are regarded as two independent sets of bosonic variables. Since $\underline{\hat{\mathbf{J}}}_{\mathrm{N}e}(\mathbf{r},\omega)$ and $\underline{\hat{\mathbf{J}}}_{\mathrm{N}m}(\mathbf{r},\omega)$ can be linearly related to $\underline{\hat{\mathbf{j}}}_{\mathrm{N}\parallel}(\mathbf{r},\omega)$ and $\underline{\hat{\mathbf{j}}}_{\mathrm{N}\perp}(\mathbf{r},\omega)$ and thus to $\underline{\hat{\mathbf{j}}}_{\mathrm{N}}(\mathbf{r},\omega)$, the variables $\hat{\mathbf{f}}_e(\mathbf{r},\omega)$ and $\hat{\mathbf{f}}_m(\mathbf{r},\omega)$ may be viewed as projective variables in the sense outlined at the end of Sec. 2.3. Since Eq. (2.115) [with Eqs. (2.116) and (2.117)] is a separation of the noise current density into a part attributed to a dielectric polarization and a part attributed to a (paramagnetic) magnetization, the quantization scheme based on Eqs. (2.115)–(2.117), with $\hat{\mathbf{f}}_e(\mathbf{r},\omega)$ and $\hat{\mathbf{f}}_m(\mathbf{r},\omega)$

being bosonic variables, may be thought of as following from the general quantization scheme in the case where magneto-electric crossing effects can be a priori excluded from consideration.

Local Limit: Other Kinds of Media

The transition to the local limit is not a unique procedure in general. Various kinds of locally responding (homogeneous) media, including non-isotropic ones, may therefore be derived as limiting cases from Eq. (2.83). To illustrate this, let us represent $\boldsymbol{\sigma}(\mathbf{k},\omega)$ as given in Eq. (2.84) in a different orthonormal basis, where the new expansion will be non-diagonal in general,

$$\boldsymbol{\sigma}(\mathbf{k},\omega) = \sum_{i,j=1}^{3} \tilde{\sigma}_{ij}(\mathbf{k},\omega)\, \tilde{\mathbf{e}}_i(\mathbf{k},\omega)\tilde{\mathbf{e}}_j^*(\mathbf{k},\omega). \tag{2.118}$$

The new basis vectors $\tilde{\mathbf{e}}_i(\mathbf{k},\omega)$ are related to the ones appearing in Eq. (2.84) by a unitary transformation,

$$\tilde{\mathbf{e}}_i(\mathbf{k},\omega) = \sum_{k=1}^{3} U_{ik}(\mathbf{k},\omega)\mathbf{e}_k(\mathbf{k},\omega), \tag{2.119}$$

$$U_{ik}(\mathbf{k},\omega) = \tilde{\mathbf{e}}_i(\mathbf{k},\omega)\cdot \mathbf{e}_k^*(\mathbf{k},\omega). \tag{2.120}$$

We may always choose the $\tilde{\mathbf{e}}_i(\mathbf{k},\omega)$ so that they are independent of $\mathbf{k}$, $\tilde{\mathbf{e}}_i(\mathbf{k},\omega)\mapsto\tilde{\mathbf{e}}_i(\omega)$. If this choice can be made such that the new expansion coefficients,

$$\tilde{\sigma}_{ij}(\mathbf{k},\omega) = \sum_{k,l=1}^{3} U_{ik}^*(\mathbf{k},\omega)\sigma_{kl}(\mathbf{k},\omega)U_{jl}(\mathbf{k},\omega), \tag{2.121}$$

may be approximately replaced under the $\mathbf{k}$-integral according to

$$\tilde{\sigma}_{ij}(\mathbf{k},\omega) \mapsto \tilde{\sigma}_{ij}(\mathbf{k}\to 0,\omega) \equiv \tilde{\sigma}_{ij}(\omega) \tag{2.122}$$

when Eq. (2.118) is inserted in Eq. (2.83), then in this way the type of locally responding (homogeneous) anisotropic medium defined by Eq. (2.73) is recovered. [Equation (2.74) is then obtained by diagonalizing $\tilde{\sigma}_{ij}(\omega)$ by means of yet another ($\mathbf{k}$-independent) unitary transformation.] Similarly, if the approximation (2.122) is generalized to include further terms of an (assumed) expansion of $\tilde{\sigma}_{ij}(\mathbf{k},\omega)$ at $\mathbf{k}=0$, then quasi-local approximations of Eq. (2.83) are generated, by inserting the truncated expansion into Eq. (2.83) and integrating term by term to yield a linear combination of various derivatives of δ-functions. In pursuing such approximation procedures—whose validity has to be examined in each case and which depends crucially on the choice of the transformation (2.119) and (2.120) (i.e., on the choice of the new basis vectors)—it must be kept in mind that any approximate form of $\boldsymbol{\sigma}(\mathbf{r},\mathbf{r}',\omega)$ so derived has to conform to all the general requirements on $\boldsymbol{Q}(\mathbf{r},\mathbf{r}',\omega)$.

In this context, let us address so-called bi-anisotropic media—the most general kind of locally responding linear media. From the above, such a medium may be viewed as corresponding to a quasi-local approximation of $\boldsymbol{\sigma}(\mathbf{r},\mathbf{r}',\omega)$ [and hence of $\boldsymbol{Q}(\mathbf{r},\mathbf{r}',\omega)$] that incorporates derivatives of δ-functions up to second order. In fact, the constitutive relations, which in classical electrodynamics, for homogeneous bi-anisotropic media, are typically given in a form such as (see, e.g., Refs. [31, 48])

$$\underline{\mathbf{P}}(\mathbf{r},\omega) = \boldsymbol{\xi}_{PE}(\omega)\cdot\underline{\mathbf{E}}(\mathbf{r},\omega) + \boldsymbol{\xi}_{PB}(\omega)\cdot\underline{\mathbf{B}}(\mathbf{r},\omega), \tag{2.123}$$

$$\underline{\mathbf{M}}(\mathbf{r},\omega) = \boldsymbol{\xi}_{ME}(\omega)\cdot\underline{\mathbf{E}}(\mathbf{r},\omega) + \boldsymbol{\xi}_{MB}(\omega)\cdot\underline{\mathbf{B}}(\mathbf{r},\omega), \tag{2.124}$$

with $\underline{\mathbf{P}}(\mathbf{r},\omega)$ and $\underline{\mathbf{M}}(\mathbf{r},\omega)$, respectively, being the polarization and magnetization fields in the frequency domain, may be equivalently given in the form of the first term of Eq. (2.8), where the appropriate conductivity tensor reads

$$\begin{aligned}\boldsymbol{Q}(\mathbf{r},\mathbf{r}',\omega) = {}& -i\omega\boldsymbol{\xi}_{PE}(\omega)\delta(\mathbf{r}-\mathbf{r}') + [\boldsymbol{\xi}_{PB}(\omega)\delta(\mathbf{r}-\mathbf{r}')]\times\overleftarrow{\boldsymbol{\nabla}}' \\ &+ \boldsymbol{\nabla}\times[\boldsymbol{\xi}_{ME}(\omega)\delta(\mathbf{r}-\mathbf{r}')] - (i\omega)^{-1}\boldsymbol{\nabla}\times[\boldsymbol{\xi}_{MB}(\omega)\delta(\mathbf{r}-\mathbf{r}')]\times\overleftarrow{\boldsymbol{\nabla}}',\end{aligned} \tag{2.125}$$

and the polarization and magnetization current densities in the frequency domain, $-i\omega\underline{\mathbf{P}}(\mathbf{r},\omega)$ and $\boldsymbol{\nabla}\times\underline{\mathbf{M}}(\mathbf{r},\omega)$, respectively, are combined into $\underline{\mathbf{j}}(\mathbf{r},\omega)$. It is not difficult to prove that the validity of the reciprocity property of $\boldsymbol{Q}(\mathbf{r},\mathbf{r}',\omega)$ as given in Eq. (2.125) is equivalent to the validity of the transposition relations $\boldsymbol{\xi}_{PE}(\omega)=\boldsymbol{\xi}^{\mathsf{T}}_{PE}(\omega)$, $\boldsymbol{\xi}_{MB}(\omega)=\boldsymbol{\xi}^{\mathsf{T}}_{MB}(\omega)$, and $\boldsymbol{\xi}_{ME}(\omega)=-\boldsymbol{\xi}^{\mathsf{T}}_{PB}(\omega)$.

If the last (magnetic-like) term on the right-hand side of Eq. (2.125) can be omitted, then one can entirely drop Eq. (2.124) and change instead Eq. (2.123) so as to read

$$\begin{aligned}\underline{\mathbf{P}}(\mathbf{r},\omega) = \boldsymbol{\xi}_{PE}(\omega)\cdot\underline{\mathbf{E}}(\mathbf{r},\omega) + \frac{1}{i\omega}\big\{&\boldsymbol{\xi}_{PB}(\omega)\cdot[\boldsymbol{\nabla}\times\underline{\mathbf{E}}(\mathbf{r},\omega)] \\ &- \boldsymbol{\nabla}\times[\boldsymbol{\xi}_{ME}(\omega)\cdot\underline{\mathbf{E}}(\mathbf{r},\omega)]\big\}.\end{aligned} \tag{2.126}$$

Introducing a third-rank tensor with Cartesian components (antisymmetric in the first two indices)

$$\zeta_{ijk}(\omega) = (i\omega)^{-1}[\xi_{PB\,il}(\omega)\,\epsilon_{jlk} + \xi_{ME\,lj}(\omega)\,\epsilon_{ilk}] \tag{2.127}$$

(ϵ_{jlk}, Levi-Civita permutation symbol; summation over indices occurring twice is understood), we may rewrite Eq. (2.126) as

$$\underline{P}_i(\mathbf{r},\omega) = \xi_{PE\,ij}\,\underline{E}_j(\mathbf{r},\omega) + \zeta_{ijk}(\omega)\partial_k\underline{E}_j(\mathbf{r},\omega), \tag{2.128}$$

which exactly corresponds to the standard form commonly used (see, e.g., Ref. [49]).

2.4.3 Spatially Dispersive Inhomogeneous Media

As already mentioned, knowledge of the medium properties, i.e., of $\boldsymbol{\sigma}(\mathbf{r},\mathbf{r}',\omega)$, is required in order to solve the eigenvalue problem (2.42) and to perform explicitly the quantization of the medium-assisted electromagnetic field—a task which, in general, cannot be accomplished in closed form. Nevertheless, to provide some analytical insight into the problem, let us consider media that combine the features of the media considered in Secs. 2.4.1 and 2.4.2 in an approximate fashion.

Model

We assume that the medium permits one to clearly distinguish between the length scales associated with spatial dispersion and inhomogeneity, with the former scale being sufficiently small as compared with the latter one. In this case, the medium can be regarded as having locally the properties of bulk material, and $\boldsymbol{\sigma}(\mathbf{r},\mathbf{r}',\omega)$ may be approximated as

$$\boldsymbol{\sigma}(\mathbf{r},\mathbf{r}',\omega) = \frac{1}{\Omega}\sum_{\mathbf{L}}\sum_{i=1}^{3}\sum_{\mathbf{k}}\sigma_{i\mathbf{Lk}}(\omega)e^{i\mathbf{k}\cdot(\mathbf{r}-\mathbf{r}')}\theta_{\mathbf{L}}(\mathbf{r})\theta_{\mathbf{L}}(\mathbf{r}')\,\mathbf{e}_{i\mathbf{Lk}}(\omega)\mathbf{e}^{*}_{i\mathbf{Lk}}(\omega), \tag{2.129}$$

from which the eigenfunctions of the associated operator are seen to be

$$\mathbf{F}_{i\mathbf{Lk}}(\mathbf{r},\omega) = \Omega^{-1/2}\theta_{\mathbf{L}}(\mathbf{r})\,e^{i\mathbf{k}\cdot\mathbf{r}}\mathbf{e}_{i\mathbf{Lk}}(\omega). \tag{2.130}$$

Here the medium is thought of as being divided into unit cells of volume Ω which form a Bravais-type lattice, the cut-off function $\theta_{\mathbf{L}}(\mathbf{r})$ is unity if $\mathbf{r}$ is in the cell of lattice vector $\mathbf{L}$ and zero otherwise, $\mathbf{e}_{i\mathbf{Lk}}(\omega)$ are, for chosen $\mathbf{L}$, $\mathbf{k}$, and ω, a triplet ($i=1,2,3$) of orthogonal unit vectors, and the wave vector $\mathbf{k}$ runs over the reciprocal lattice. Note that, for each cell $\mathbf{L}$,

$$\boldsymbol{\sigma}_{\mathbf{Lk}}(\omega) = \sum_{i=1}^{3}\sigma_{i\mathbf{Lk}}(\omega)\mathbf{e}_{i\mathbf{Lk}}(\omega)\mathbf{e}^{*}_{i\mathbf{Lk}}(\omega) \tag{2.131}$$

corresponds to the diagonal form in the $(\mathbf{k},\omega)$ domain of $\boldsymbol{\sigma}(\mathbf{r},\mathbf{r}',\omega)$ for bulk material [cf. Eq. (2.84)].

The main features of $\boldsymbol{\sigma}(\mathbf{r},\mathbf{r}',\omega)$ as given in Eq. (2.129) can be summarized as follows. (i) $\boldsymbol{\sigma}(\mathbf{r},\mathbf{r}',\omega)$ is zero whenever $\mathbf{r}$ and $\mathbf{r}'$ are not in the same cell, so that $\Omega^{1/3}$ determines the length scale on which spatial dispersion is at most observed. (ii) The dependence on $\mathbf{L}$ of $\boldsymbol{\sigma}_{\mathbf{L}\mathbf{k}}(\omega)$ [Eq. (2.131)] for an inhomogeneous medium introduces an $\mathbf{L}$-dependence into Eq. (2.129) which should be sufficiently weak, so that noticeable violations of the translational invariance of $\boldsymbol{\sigma}(\mathbf{r},\mathbf{r}',\omega)$ may occur only on a length scale that is large compared with $\Omega^{1/3}$. Needless to say that the main features do not essentially change if $\theta_{\mathbf{L}}(\mathbf{r})$ is replaced with another—but qualitatively similar—cut-off function.

Let us denote by $\mathbf{L}(\mathbf{r})$ the particular lattice vector whose cell contains the point $\mathbf{r}$, so that $\mathbf{L}(\mathbf{r})$ plays the role of a coarse-grained position variable. With the notations $\theta_{\mathbf{L}(\mathbf{r})}(\mathbf{r}')\mapsto\theta[\mathbf{L}(\mathbf{r}),\mathbf{r}']$ and $\boldsymbol{\sigma}_{\mathbf{L}(\mathbf{r})\mathbf{k}}(\omega)\mapsto\boldsymbol{\sigma}_{\mathbf{k}}[\mathbf{L}(\mathbf{r}),\omega]$, Eq. (2.129) together with Eq. (2.131) can be rewritten as

$$\boldsymbol{\sigma}(\mathbf{r},\mathbf{r}',\omega)=\theta[\mathbf{L}(\mathbf{r}'),\mathbf{r}]\boldsymbol{\sigma}[\mathbf{L}(\mathbf{r}'),\mathbf{r}-\mathbf{r}',\omega], \tag{2.132}$$

with

$$\boldsymbol{\sigma}[\mathbf{L}(\mathbf{r}'),\mathbf{r}-\mathbf{r}',\omega]=\frac{1}{\Omega}\sum_{\mathbf{k}}\boldsymbol{\sigma}_{\mathbf{k}}[\mathbf{L}(\mathbf{r}'),\omega]e^{i\mathbf{k}\cdot(\mathbf{r}-\mathbf{r}')}. \tag{2.133}$$

Note that for arbitrary (continuous) values $\mathbf{s}$, the function $\theta(\mathbf{s},\mathbf{r})$ can be regarded as being symmetric. Using Eq. (2.130), we find that Eq. (2.46) takes the form

$$\boldsymbol{K}(\mathbf{r},\mathbf{r}',\omega)=\frac{\theta[\mathbf{L}(\mathbf{r}'),\mathbf{r}]}{\Omega}\sum_{\mathbf{k}}\boldsymbol{K}_{\mathbf{k}}[\mathbf{L}(\mathbf{r}'),\omega]e^{i\mathbf{k}\cdot(\mathbf{r}-\mathbf{r}')}, \tag{2.134}$$

where

$$\boldsymbol{K}_{\mathbf{k}}[\mathbf{L}(\mathbf{r}),\omega]=\sum_{i=1}^{3}\sigma_{i\mathbf{k}}^{1/2}[\mathbf{L}(\mathbf{r}),\omega]\,\mathbf{e}_{i\mathbf{k}}[\mathbf{L}(\mathbf{r}),\omega]\mathbf{e}^{*}_{i\mathbf{k}}[\mathbf{L}(\mathbf{r}),\omega] \tag{2.135}$$

$\{\sigma_{i\mathbf{L}(\mathbf{r})\mathbf{k}}(\omega)\mapsto\sigma_{i\mathbf{k}}[\mathbf{L}(\mathbf{r}),\omega],\ \mathbf{e}_{i\mathbf{L}(\mathbf{r})\mathbf{k}}(\omega)\mapsto\mathbf{e}_{i\mathbf{k}}[\mathbf{L}(\mathbf{r}),\omega]\}$.

It can be shown that Eq. (2.132) [with Eq. (2.133)] indeed contains (and, in a sense, interpolates) the two limiting cases studied in Secs. 2.4.1 and 2.4.2. For the proof, we observe that in the case of negligible spatial dispersion, the cell size can be shrunk to zero, $\Omega\to 0$, so that the lattice vectors take on continuous values, $\mathbf{L}(\mathbf{r})\to\mathbf{r}$. As the lattice becomes finer and finer, the reciprocal lattice becomes more and more coarse, and, for $\mathbf{r}$ and $\mathbf{r}'$ unequal but in the same cell, all the points of the reciprocal lattice with $\mathbf{k}\neq 0$ give rise to rapidly oscillating terms in Eq. (2.133). In the limit $\Omega\to 0$, these terms oscillate infinitely rapidly and average to zero (when applying the operator associated with Eq. (2.132) [with Eq. (2.133)] to any reasonable function), so that they may be set equal to zero. Taking also into account that $\theta[\mathbf{L}(\mathbf{r}'),\mathbf{r}]/\Omega \to \delta(\mathbf{r}-\mathbf{r}')$ in this limit, we see that Eq. (2.132) [with Eq. (2.133)] indeed approaches Eq. (2.73) for vanishing spatial dispersion [note the correspondences $\sigma_{i\mathbf{k}=0}[\mathbf{L}(\mathbf{r})=\mathbf{r},\omega]=\sigma_i(\mathbf{r},\omega)$ and $\mathbf{e}_{i\mathbf{k}=0}[\mathbf{L}(\mathbf{r})=\mathbf{r},\omega] =\mathbf{e}_i(\mathbf{r},\omega)$].

On the other hand, in the limiting case of an infinitely extended homogeneous medium, there is no $\mathbf{L}$-dependence of the medium properties so that we are free to increase the cell size indefinitely, $\Omega\to\infty$. Consequently, we may let $\theta[\mathbf{L}(\mathbf{r}'),\mathbf{r}]\to 1$ in Eq. (2.132) and $\boldsymbol{\sigma}_{\mathbf{k}}[\mathbf{L}(\mathbf{r}),\omega]\to\boldsymbol{\sigma}(\mathbf{k},\omega)$, $\Omega^{-1}\sum_{\mathbf{k}}\to(2\pi)^{-3}\int \mathrm{d}^3k$ in Eq. (2.133), which reveals that Eq. (2.132) [with Eq. (2.133)] approaches Eq. (2.83) as expected.

Magnetodielectric Media

To quantize the electromagnetic field in an inhomogeneous magnetodielectric medium specified in terms of $\varepsilon(\mathbf{r},\omega)$ and $\kappa(\mathbf{r},\omega)=\mu^{-1}(\mathbf{r},\omega)$, let us consider a medium that is both sufficiently weakly inhomogeneous and sufficiently weakly spatially dispersive, so that Ω in Eq. (2.133) can be chosen on a scale intermediate between the scales of spatial dispersion and inhomogeneity. We may then approximately let $\mathbf{L}(\mathbf{r})$ be a continuous variable, $\mathbf{L}(\mathbf{r})\to\mathbf{r}$ in Eq. (2.132), and yet, at the same time, approximately treat the $\mathbf{k}$-sum in Eq. (2.133) as an integral, so that Eq. (2.132) [with Eq. (2.133)] approximates to $[\theta[\mathbf{L}(\mathbf{r}'),\mathbf{r}]\to\theta(\mathbf{r}',\mathbf{r})]$

$$\boldsymbol{\sigma}(\mathbf{r},\mathbf{r}',\omega)=\frac{\theta(\mathbf{r}',\mathbf{r})}{(2\pi)^3}\int \mathrm{d}^3k\,\boldsymbol{\sigma}(\mathbf{r}',\mathbf{k},\omega)e^{i\mathbf{k}\cdot(\mathbf{r}-\mathbf{r}')}. \tag{2.136}$$

For a medium that is locally of the type described by Eq. (2.95), we may set

$$\boldsymbol{\sigma}(\mathbf{r},\mathbf{k},\omega)=\sigma_{\|}(\mathbf{r},k,\omega)\boldsymbol{I}-\mathbf{k}\times\gamma(\mathbf{r},k,\omega)\boldsymbol{I}\times\mathbf{k}, \tag{2.137}$$

where

$$\gamma(\mathbf{r},k,\omega)=[\sigma_{\perp}(\mathbf{r},k,\omega)-\sigma_{\|}(\mathbf{r},k,\omega)]/k^2>0. \tag{2.138}$$

Assuming that in the $\mathbf{k}$-integral in Eq. (2.136), $\sigma_{\|}(\mathbf{r},k,\omega)$ and $\gamma(\mathbf{r},k,\omega)$ may be approximated, respectively, by well-defined (and unique) long-wavelength limits $\sigma_\parallel(\mathbf{r},\omega) = \lim_{k\to 0}\sigma_\parallel(\mathbf{r},k,\omega)$ and $\gamma(\mathbf{r},\omega) = \lim_{k\to 0}\gamma(\mathbf{r},k,\omega)$, the cut-off function $\theta(\mathbf{r}',\mathbf{r})$ has—due to the rapid oscillations of the exponential for large $|\mathbf{r}-\mathbf{r}'|$—no effect [with regard to an application of the operator associated with $\boldsymbol{\sigma}(\mathbf{r},\mathbf{r}',\omega)$ from Eq. (2.136)] and can be dropped, and we obtain, as a generalization of Eq. (2.103),

$$\boldsymbol{\sigma}(\mathbf{r},\mathbf{r}',\omega) = \sigma_\parallel(\mathbf{r}',\omega)\boldsymbol{I}\delta(\mathbf{r}-\mathbf{r}') - \boldsymbol{\nabla}\times[\gamma(\mathbf{r}',\omega)\boldsymbol{I}\delta(\mathbf{r}-\mathbf{r}')]\times\overleftarrow{\boldsymbol{\nabla}}'. \tag{2.139}$$

With the identifications

$$\sigma_\parallel(\mathbf{r},\omega) = \varepsilon_0\omega\,\mathrm{Im}\,\varepsilon(\mathbf{r},\omega), \tag{2.140}$$
$$\gamma(\mathbf{r},\omega) = -\kappa_0\mathrm{Im}\,\kappa(\mathbf{r},\omega)/\omega \tag{2.141}$$

[cf. Eqs. (2.108) and (2.109)], Eqs. (2.104) and (2.105) generalize to

$$\begin{aligned}\boldsymbol{Q}(\mathbf{r},\mathbf{r}',\omega) = {}&-i\varepsilon_0\omega\,[\varepsilon(\mathbf{r}',\omega)-1]\boldsymbol{I}\delta(\mathbf{r}-\mathbf{r}')\\ &-\frac{1}{i\omega}\boldsymbol{\nabla}\times\{\kappa_0[1-\kappa(\mathbf{r}',\omega)]\boldsymbol{I}\delta(\mathbf{r}-\mathbf{r}')\}\times\overleftarrow{\boldsymbol{\nabla}}'\end{aligned} \tag{2.142}$$

and

$$\underline{\hat{\mathbf{j}}}(\mathbf{r},\omega) = -i\varepsilon_0\omega[\varepsilon(\mathbf{r},\omega)-1]\underline{\hat{\mathbf{E}}}(\mathbf{r},\omega)+\kappa_0\boldsymbol{\nabla}\times\{[1-\kappa(\mathbf{r},\omega)]\underline{\hat{\mathbf{B}}}(\mathbf{r},\omega)\}+\underline{\hat{\mathbf{j}}}_{\mathrm{N}}(\mathbf{r},\omega), \tag{2.143}$$

respectively.

Unfortunately, Eq. (2.110) does not generalize to

$$\boldsymbol{K}'(\mathbf{r},\mathbf{r}',\omega) = \sigma_\parallel^{1/2}(\mathbf{r}',\omega)\boldsymbol{I}\delta(\mathbf{r}-\mathbf{r}')\mp i\gamma^{1/2}(\mathbf{r}',\omega)\boldsymbol{\nabla}\times\boldsymbol{I}\delta(\mathbf{r}-\mathbf{r}')\quad\text{(wrong!)}, \tag{2.144}$$

as could have been suspected. Indeed, straightforward calculation shows that, for spatially varying permittivity and permeability, the kernel (2.144) does not solve Eq. (2.33) [with $\boldsymbol{\sigma}(\mathbf{r},\mathbf{r}',\omega)$ as given in Eq. (2.139)], which implies that $\underline{\hat{\mathbf{j}}}_{\mathrm{N}}(\mathbf{r},\omega)$ cannot be related to the variables $\hat{\mathbf{f}}(\mathbf{r},\omega)$ as in Eq. (2.111), with $\varepsilon(\omega)$ and $\kappa(\omega)$ being simply replaced with their inhomogeneous counterparts $\varepsilon(\mathbf{r},\omega)$ and $\kappa(\mathbf{r},\omega)$, respectively. In order to obtain an explicit expression for the kernel $\boldsymbol{K}(\mathbf{r},\mathbf{r}',\omega)$ required in Eq. (2.32), one has instead to return to Eq. (2.33) and solve it with $\boldsymbol{\sigma}(\mathbf{r},\mathbf{r}',\omega)$ from Eq. (2.139)—a problem that is, however, very difficult to solve in general. Although this does not at all limit the practical applicability of the theory [since all one typically has to know about $\boldsymbol{K}(\mathbf{r},\mathbf{r}',\omega)$ is that it satisfies its defining equation (2.33) for the chosen conductivity (2.142)], it may be useful to have at hand at least an approximate form for weak inhomogeneity, such as (see Ref. [R10] for the derivation)

$$\boldsymbol{K}(\mathbf{r},\mathbf{r}',\omega) = \sigma_{\|}^{1/2}(\omega)\boldsymbol{I}\delta(\mathbf{r}-\mathbf{r}') \mp i\gamma^{1/2}(\omega)\boldsymbol{\nabla}\times\boldsymbol{I}\delta(\mathbf{r}-\mathbf{r}')$$
$$+\tfrac{1}{2}[\sigma_{\|}(\mathbf{r},\omega)-\sigma_{\|}(\omega)]\boldsymbol{M}_0(\mathbf{r},\mathbf{r}',\omega)+\tfrac{1}{2}\boldsymbol{\nabla}\times\big\{[\gamma(\mathbf{r},\omega)-\gamma(\omega)]\boldsymbol{\nabla}\times\boldsymbol{M}_0(\mathbf{r},\mathbf{r}',\omega)\big\}, \tag{2.145}$$

where

$$\begin{aligned}\boldsymbol{M}_0(\mathbf{r},\mathbf{r}',\omega) &= \sigma_{\|}^{-1/2}(\omega)\boldsymbol{I}\delta(\mathbf{r}-\mathbf{r}') \pm i\gamma^{-1/2}(\omega)\boldsymbol{\nabla}\times m_0(\mathbf{r},\mathbf{r}',\omega)\boldsymbol{I}\\ &\quad + \sigma_{\|}^{-1/2}(\omega)\boldsymbol{\nabla}\times m_0(\mathbf{r},\mathbf{r}',\omega)\boldsymbol{I}\times\overleftarrow{\boldsymbol{\nabla}}',\end{aligned} \tag{2.146}$$

with $m_0(\mathbf{r},\mathbf{r}',\omega) = -(4\pi|\mathbf{r}-\mathbf{r}'|)^{-1}\,e^{-|\mathbf{r}-\mathbf{r}'|/\alpha(\omega)}$, $\alpha(\omega) = [\gamma(\omega)/\sigma_{\|}(\omega)]^{1/2} > 0$. If the lack of exact knowledge of $\boldsymbol{K}(\mathbf{r},\mathbf{r}',\omega)$ really happens to be an obstacle in an application, one can alternatively resort to the approach on the basis of Eqs. (2.115)–(2.117), by simply replacing therein $\varepsilon(\omega)$ and $\kappa(\omega)$ by $\varepsilon(\mathbf{r},\omega)$ and $\kappa(\mathbf{r},\omega)$, respectively.

2.5 Extension to Amplifying Media

While an unperturbed medium left to its own resources is absorbing in the whole frequency range, a medium that has been brought out of equilibrium may act, in a certain frequency range, as an amplifier until after some relaxation time it will have reached the equilibrium state again. However, when—by implementation of a suitable energy supply mechanism—a medium is pumped in such a way that an (externally controlled) quasi-stationary regime is established and maintained for a sufficiently long time, one may describe the effect of the amplifying medium by assigning to it a conductivity tensor and treat it in close analogy to the case of absorbing media. The properties of such an amplifying medium are thereby regarded as being fixed—an assumption which is of course only justified in a time interval within which significant changes of these properties can be ignored (for the purpose at hand). Clearly, the conductivity tensor that describes the linear response of an amplifying medium can be expected to have all the properties known from an absorbing medium, except that its real part is no longer the kernel of a positive-definite integral operator according to Eq. (2.12). In the following we show that, under appropriate conditions, it is possible to generalize the quantization scheme to allow also for amplifying media.

If, within some frequency interval, $\boldsymbol{\sigma}(\mathbf{r},\mathbf{r}',\omega)$ does not correspond to a positive definite integral operator so that the integral in the inequality (2.12) becomes negative for suitably chosen (quadratically integrable) functions $\mathbf{v}(\mathbf{r})$, linear amplification is possible in this frequency interval. It is not difficult to prove that the equations given in Secs. 2.1 and 2.2 up to (and including) Eq. (2.29) can be maintained even if the positivity condition (2.12) is abandoned, provided that nevertheless (i) the Green tensor retains its analytic properties and (ii) $\boldsymbol{\rho}(\mathbf{r},\mathbf{r}',\omega)$ continues to exist (a detailed discussion of these conditions is given below). However, if the condition (2.12) is abandoned, it is no longer possible to put the Hamiltonian Eq. (2.27) in the diagonal form (2.30) by use of the transformation (2.32) with $\hat{\mathbf{f}}(\mathbf{r},\omega)$ being a bosonic field. The reason is that Eq. (2.33) is inconsistent if $\boldsymbol{\sigma}(\mathbf{r},\mathbf{r}',\omega)$ is not the kernel of a positive definite integral operator, so that in this case no valid kernel $\boldsymbol{K}(\mathbf{r},\mathbf{r}',\omega)$ exists. Consequently, Eq. (2.30) would fail to be an equivalent representation of Eq. (2.27), if $\hat{\mathbf{f}}(\mathbf{r},\omega)$ were regarded as being a bosonic field satisfying the commutation relation (2.31).

In order to include in the quantization scheme (linearly) amplifying media nevertheless, we note that, although Eq. (2.33) cannot be satisfied anymore so that Eqs. (2.30)–(2.31) do not apply either, Eqs. (2.42)–(2.45) remain valid, with $\sigma(\alpha,\omega)$ not being restricted to positive values anymore. Thus, we may use Eqs. (2.42)–(2.44) to expand $\boldsymbol{\rho}(\mathbf{r},\mathbf{r}',\omega)$ as

$$\boldsymbol{\rho}(\mathbf{r},\mathbf{r}',\omega) = \int \mathrm{d}\alpha\, \sigma^{-1}(\alpha,\omega)\mathbf{F}(\alpha,\mathbf{r},\omega)\mathbf{F}^{*}(\alpha,\mathbf{r}',\omega), \tag{2.147}$$

which enables us to rewrite the Hamiltonian (2.27) as

$$\hat{H} = \int_0^\infty \mathrm{d}\omega\, \hbar\omega \int \mathrm{d}\alpha\, \mathrm{sgn}\, \sigma(\alpha,\omega)\hat{\tilde{g}}^\dagger(\alpha,\omega)\hat{\tilde{g}}(\alpha,\omega) \tag{2.148}$$

where

$$\hat{\tilde{g}}(\alpha,\omega) = \left(\frac{\hbar\omega}{\pi}\right)^{-\frac{1}{2}} |\sigma(\alpha,\omega)|^{-\frac{1}{2}} \int \mathrm{d}^3 r\, \mathbf{F}^{*}(\alpha,\mathbf{r},\omega)\cdot \hat{\underline{\mathbf{j}}}_{\mathrm{N}}(\mathbf{r},\omega). \tag{2.149}$$

With the help of Eqs. (2.25), (2.42), and (2.44), it is not difficult to see that

$$\left[\hat{\tilde{g}}(\alpha,\omega), \hat{\tilde{g}}^\dagger(\alpha',\omega')\right] = \mathrm{sgn}\, \sigma(\alpha,\omega)\, \delta(\alpha-\alpha')\delta(\omega-\omega'). \tag{2.150}$$

The $\hat{\tilde{g}}(\alpha,\omega)$ may be viewed as generalizations of the natural variables considered in Sec. 2.3, as they coincide with the latter in the purely absorbing case where $\operatorname{sgn}\sigma(\alpha,\omega)=1$. Inversion of Eq. (2.149) by means of Eq. (2.43) yields

$$\hat{\underline{\mathbf{j}}}_{\mathrm{N}}(\mathbf{r},\omega)=\left(\frac{\hbar\omega}{\pi}\right)^{\frac{1}{2}}\int \mathrm{d}\alpha\,|\sigma(\alpha,\omega)|^{\frac{1}{2}}\,\mathbf{F}(\alpha,\mathbf{r},\omega)\hat{\tilde{g}}(\alpha,\omega) \tag{2.151}$$

as the generalization of Eq. (2.53).

The commutation relation (2.150) shows that $\hat{\tilde{g}}(\alpha,\omega)$ $[\hat{\tilde{g}}^\dagger(\alpha,\omega)]$ is a bosonic annihilation (creation) operator for positive eigenvalues $\sigma(\alpha,\omega)$ whereas for negative ones, $\hat{\tilde{g}}(\alpha,\omega)$ $[\hat{\tilde{g}}^\dagger(\alpha,\omega)]$ is a creation (annihilation) operator. It makes therefore sense to rename the operators according to this behavior. Thus, denoting in each of the two cases $\operatorname{sgn}\sigma(\alpha,\omega)=\pm 1$ the respective annihilation operator by $\hat{b}(\alpha,\omega)$ and the respective creation operator by $\hat{b}^\dagger(\alpha,\omega)$, we can rewrite Eqs. (2.148), (2.150), and (2.151) as

$$\hat{H}=\int_0^\infty \mathrm{d}\omega\int_{(+)}\mathrm{d}\alpha\,\hbar\omega\hat{b}^\dagger(\alpha,\omega)\hat{b}(\alpha,\omega)-\int_0^\infty \mathrm{d}\omega\int_{(-)}\mathrm{d}\alpha\,\hbar\omega\hat{b}(\alpha,\omega)\hat{b}^\dagger(\alpha,\omega), \tag{2.152}$$

$$\left[\hat{b}(\alpha,\omega),\hat{b}^\dagger(\alpha',\omega')\right]=\delta(\alpha-\alpha')\delta(\omega-\omega'), \tag{2.153}$$

and

$$\hat{\underline{\mathbf{j}}}_{\mathrm{N}}(\mathbf{r},\omega)=\left(\frac{\hbar\omega}{\pi}\right)^{\frac{1}{2}}\biggl\{\int_{(+)}\mathrm{d}\alpha\,\sigma^{\frac{1}{2}}(\alpha,\omega)\,\mathbf{F}(\alpha,\mathbf{r},\omega)\hat{b}(\alpha,\omega) + \int_{(-)}\mathrm{d}\alpha\,[-\sigma(\alpha,\omega)]^{\frac{1}{2}}\,\mathbf{F}(\alpha,\mathbf{r},\omega)\hat{b}^\dagger(\alpha,\omega)\biggr\}, \tag{2.154}$$

respectively, where the notation $\int_{(\pm)}\mathrm{d}\alpha\cdots$ means that the integration extends over those values for which $\operatorname{sgn}\sigma(\alpha,\omega)=\pm 1$. Note that the ranges of integration depend on the chosen frequency in general. It may be convenient to change to normal order the second term in Eq. (2.152), $\hat{b}(\alpha,\omega)\hat{b}^\dagger(\alpha,\omega)$ $\mapsto :\ \hat{b}(\alpha,\omega)\hat{b}^\dagger(\alpha,\omega)\ := \hat{b}^\dagger(\alpha,\omega)\hat{b}(\alpha,\omega)$, i.e., to replace the Hamiltonian in Eq. (2.152) with

$$\hat{H}=\int_0^\infty \mathrm{d}\omega\,\hbar\omega\int \mathrm{d}\alpha\,\operatorname{sgn}\sigma(\alpha,\omega)\hat{b}^\dagger(\alpha,\omega)\hat{b}(\alpha,\omega), \tag{2.155}$$

which differs from the Hamiltonian in Eq. (2.152) by an (infinite but) irrelevant c-number. Since the state space of the system is to be constructed by means of the bosonic variables $\hat{b}(\alpha,\omega)$ and $\hat{b}^\dagger(\alpha,\omega)$, the use of Eq. (2.155) in place of Eq. (2.152) is equivalent to a redefinition (renormalization) of the zero of energy by the condition of absence of quanta (vacuum state). It should be stressed that the temporal evolution of the variables $\hat{b}(\alpha,\omega)$ and $\hat{b}^\dagger(\alpha,\omega)$ that follows from Eq. (2.155) [together with Eq. (2.153)] is sensitive to the sign of $\sigma(\alpha,\omega)$ in just such a way that Eq. (2.154) always represents the positive-frequency part of the noise current density, as required.

From Eq. (2.155) it is seen that there is a continuum of negative energy eigenvalues in the case of amplification, in addition to the positive-energy continuum associated with absorption. Thus, the vacuum state can no longer be said to be the ground state of the system—there is in fact no ground state as the continuum stretches down to $-\infty$. This somewhat unpleasant feature is due to the fact that the pump mechanism which prepares the medium to act, in some frequency interval, as an amplifier is not dynamically included in the theory. Clearly, the approximation to treat the effect of pumping within the framework of linear response theory breaks down when states with large negative energies are significantly involved.

By means of the transformation

$$\begin{aligned}\hat{\hat{\mathbf{f}}}(\mathbf{r},\omega) &= \int \mathrm{d}\alpha\,\mathbf{F}(\alpha,\mathbf{r},\omega)\hat{\hat{g}}(\alpha,\omega)\\ &= \int_{(+)} \mathrm{d}\alpha\,\mathbf{F}(\alpha,\mathbf{r},\omega)\hat{b}(\alpha,\omega) + \int_{(-)} \mathrm{d}\alpha\,\mathbf{F}(\alpha,\mathbf{r},\omega)\hat{b}^\dagger(\alpha,\omega),\end{aligned} \tag{2.156}$$

vectorial field variables $\hat{\hat{\mathbf{f}}}(\mathbf{r},\omega)$ can be introduced, which can be viewed as generalizations of the variables $\hat{\mathbf{f}}(\mathbf{r},\omega)$ introduced in Eq. (2.32) for the case of purely absorbing media. Employing Eq. (2.44) to invert (the first equation in) Eq. (2.156) and inserting the result in Eq. (2.151), we obtain the generalization of Eq. (2.32) [$\hat{\mathbf{f}}(\mathbf{r},\omega) \mapsto \hat{\hat{\mathbf{f}}}(\mathbf{r},\omega)$], from which we can read off, by comparison with Eq. (2.32), the generalization of the kernel $\boldsymbol{K}(\mathbf{r},\mathbf{r}',\omega)$, in the form of its eigenfunction expansion. Not surprisingly, it is given by Eq. (2.46) with $\sigma(\alpha,\omega)$ being replaced by $|\sigma(\alpha,\omega)|$. (It is of course possible to consider equivalent kernels just as in the case of purely absorbing media.) Unfortunately, the variables $\hat{\hat{\mathbf{f}}}(\mathbf{r},\omega)$ do not diagonalize the Hamiltonian in general, and are thus less useful than in the case of purely absorbing media.

From Eqs. (2.150) and (2.156), it follows that

$$[\hat{\tilde{\mathbf{f}}}(\mathbf{r},\omega),\hat{\tilde{\mathbf{f}}}^\dagger(\mathbf{r}',\omega')]=\delta(\omega-\omega')\int \mathrm{d}\alpha\,\mathrm{sgn}\,\sigma(\alpha,\omega)\,\mathbf{F}(\alpha,\mathbf{r},\omega)\mathbf{F}^*(\alpha,\mathbf{r}',\omega). \tag{2.157}$$

The integral appearing on the right-hand side is the kernel of a parity-type operator, which reduces to the unit operator in the case of purely absorbing media, as it should be [cf. Eqs. (2.31) and (2.43)].

A noteworthy simplification occurs if the medium response is strictly local—an assumption that is typically made in the study of amplifying media. In this case, the $\hat{\tilde{\mathbf{f}}}(\mathbf{r},\omega)$ are related to the $\hat{\tilde{g}}(\alpha,\omega)$ $[\alpha\mapsto(i,\mathbf{r})]$ in a very simple way just as in the case of purely absorbing media that respond locally [since the eigenfunctions $\mathbf{F}(\alpha,\mathbf{r},\omega)$ are spatially localized in this case, see Sec. 2.4.1]. Focusing, for simplicity, on isotropic media, we may then rewrite Eqs. (2.151) and (2.148), respectively, as

$$\hat{\underline{\mathbf{j}}}_{\mathrm{N}}(\mathbf{r},\omega)=\left(\frac{\hbar\omega}{\pi}\right)^{\frac{1}{2}}|\sigma(\mathbf{r},\omega)|^{\frac{1}{2}}\,\hat{\tilde{\mathbf{f}}}(\mathbf{r},\omega), \tag{2.158}$$

and

$$\hat{H}=\int_0^\infty \mathrm{d}\omega\,\hbar\omega\int \mathrm{d}^3r\,\mathrm{sgn}\,\sigma(\mathbf{r},\omega)\hat{\tilde{\mathbf{f}}}^\dagger(\mathbf{r},\omega)\cdot\hat{\tilde{\mathbf{f}}}(\mathbf{r},\omega), \tag{2.159}$$

and (2.157) simplifies to

$$\left[\hat{\tilde{\mathbf{f}}}(\mathbf{r},\omega),\hat{\tilde{\mathbf{f}}}^\dagger(\mathbf{r}',\omega')\right]=\mathrm{sgn}\,\sigma(\mathbf{r},\omega)\,\boldsymbol{I}\delta(\mathbf{r}-\mathbf{r}')\delta(\omega-\omega'). \tag{2.160}$$

Now we can switch to genuine bosonic variables by renaming $\hat{\tilde{\mathbf{f}}}(\mathbf{r},\omega)$ as $\hat{\mathbf{b}}(\mathbf{r},\omega)$ and $\hat{\mathbf{b}}^\dagger(\mathbf{r},\omega)$ for $\mathrm{sgn}\,\sigma(\mathbf{r},\omega)=1$ and $\mathrm{sgn}\,\sigma(\mathbf{r},\omega)=-1$, respectively, so that Eq. (2.160) changes to

$$\left[\hat{\mathbf{b}}(\mathbf{r},\omega),\hat{\mathbf{b}}^\dagger(\mathbf{r}',\omega')\right]=\boldsymbol{I}\delta(\mathbf{r}-\mathbf{r}')\delta(\omega-\omega'). \tag{2.161}$$

The noise current density in Eq. (2.154) and the Hamiltonian in Eq. (2.155), respectively, can then be expressed in terms of $\hat{\mathbf{b}}(\mathbf{r},\omega)$ and $\hat{\mathbf{b}}^\dagger(\mathbf{r},\omega)$ as

$$\begin{aligned}\hat{\underline{\mathbf{j}}}_{\mathrm{N}}(\mathbf{r},\omega)=\left(\frac{\hbar\omega}{\pi}\right)^{\frac{1}{2}}\Big\{&\theta[\sigma(\mathbf{r},\omega)]\sigma^{\frac{1}{2}}(\mathbf{r},\omega)\,\hat{\mathbf{b}}(\mathbf{r},\omega)\\ &+\theta[-\sigma(\mathbf{r},\omega)][-\sigma(\mathbf{r},\omega)]^{\frac{1}{2}}\,\hat{\mathbf{b}}^\dagger(\mathbf{r},\omega)\Big\}\end{aligned} \tag{2.162}$$

[$\theta(x)$, unit step function] and

$$\hat{H} = \int_0^\infty \mathrm{d}\omega\, \hbar\omega \int \mathrm{d}^3r\, \operatorname{sgn}\sigma(\mathbf{r},\omega)\hat{\mathbf{b}}^\dagger(\mathbf{r},\omega)\cdot\hat{\mathbf{b}}(\mathbf{r},\omega). \tag{2.163}$$

Taking into account that $\sigma(\mathbf{r},\omega)$ can be related to the imaginary part of the dielectric permittivity according to Eq. (2.140) [$\sigma(\mathbf{r},\omega) = \sigma_\parallel(\mathbf{r},\omega)$], we find that Eq. (2.162) is nothing but the equation suggested in Ref. [44] for the case of isotropic, locally and linearly responding dielectrics that also allow for amplification. Note that, as already mentioned for the more general case of Eq. (2.154), both the term associated with $\hat{\mathbf{b}}(\mathbf{r},\omega)$ and the term associated with $\hat{\mathbf{b}}^\dagger(\mathbf{r},\omega)$ in Eq. (2.162) give rise to positive-frequency parts of the noise current density.

Range of Validity

It remains to specify in more detail the conditions that must be satisfied to apply the quantization scheme to amplifying media. Let us first consider the question as to whether the Green tensor $\boldsymbol{G}(\mathbf{r},\mathbf{r}',\omega)$ remains analytic in the upper complex ω half-plane if linear amplification is allowed for. As remarked at the end of Sec.2.1, in the case of absorbing media solutions to the source-free macroscopic Maxwell equations [i. e., solutions to the homogeneous version of Eq. (2.16)] for real frequency ω can be ruled out because of their divergent spatial behavior. The same is obviously 'even more' true for frequencies in the upper complex ω half-plane. Since the existence of permissible solutions to the source-free equations is known to manifest itself mathematically in the form of singularities of the Green tensor, only the lower complex ω half-plane is a possible location of such singularities in the case of absorbing media. (In this context, values of the Green tensor in the lower frequency half-plane are defined—if at all possible—only through analytical continuation from the upper half-plane.) In contrast, for linearly amplifying media it may happen that permissible solutions to the source-free macroscopic Maxwell equations exist even if ω is chosen in the upper complex ω half-plane. In this case, singularities of the Green tensor in the upper complex ω half-plane would exist, which would invalidate the proof of the commutation relation in Eq. (2.24).

It is not difficult to imagine that such 'unwanted' solutions to the source-free macroscopic Maxwell equations could arise, e.g., if waves were allowed to propagate through an infinitely extended region of amplification. Since such regions do not exist in practice, their necessary exclusion from consideration is of no practical relevance. More importantly, the same problem of 'infinite amplification length' can also occur in the case of a region of amplification of finite extension, if waves can pass through the region repeatedly (due to multiple reflections) with a net gain per round-trip. Therefore, setups where an amplifying medium is part of an arrangement of bodies that act as a high-Q resonator must possibly be excluded from consideration. In such cases, the Green tensor fails to be a causal function in the sense of linear response theory. Of course, what breaks down is not really the principle of causality but the very concept of linear amplification—as fields with higher and higher energy develop, the non-linear dynamics can no longer be disregarded. In fact, there is no need for extra criteria to exclude from consideration setups that would mathematically support 'unwanted' solutions—such setups are already excluded implicitly by the assumption that the approximate concept of linear amplification is applicable. Hence, by this basic assumption, the Green tensor is forced to be analytic in the upper complex ω half-plane, and all the other important properties of the Green tensor (in particular, its high- and low-frequency behavior and its decay behavior for large difference of the spatial arguments) are the same as in the case of absorbing media.

Next, let us answer the question of the existence of the inverse of the integral operator associated with $\boldsymbol{\sigma}(\mathbf{r},\mathbf{r}',\omega)$ in the case of linear amplification, i.e., the question of the existence of the kernel $\boldsymbol{\rho}(\mathbf{r},\mathbf{r}',\omega)$ according to Eq. (2.28). Obviously, $\boldsymbol{\rho}(\mathbf{r},\mathbf{r}',\omega)$ would fail to exist if eigenvalues of the operator associated with $\boldsymbol{\sigma}(\mathbf{r},\mathbf{r}',\omega)$ were exactly equal to zero. Since amplification is limited to a certain frequency range, and since frequency is a continuous variable, each eigenvalue $\sigma(\alpha,\omega)$ on the negative side of the eigenvalue spectrum can be made to move to the positive side by tuning the frequency, so that zero eigenvalues are possible. However, it should be emphasized that in the case of a continuous spectrum this problem is in fact harmless. It is generally true that zero occurring as a continuum eigenvalue does not really preclude the inversion of (Hermitian) operators. A familiar example is provided by the free-particle Schrödinger equation considered in the whole space. There, the continuum of δ-normalizable (plane-wave) eigenfunctions includes the (spatially constant) zero-energy eigenfunction, but this does not imply the nonexistence of the inverse of the free-particle Hamiltonian but merely the unboundedness of the inverse operator. Although there are good reasons to believe that in practice the eigenvalues $\sigma(\alpha,\omega)$ form a continuous spectrum, it seems advisable to have also a method in store to handle discrete zero eigenvalues, recalling that the calculation of physical quantities we are dealing with generally involves space and frequency integrations. Therefore, as long as an expression for such a quantity remains well-defined as a whole, it does not really matter if $\boldsymbol{\rho}(\mathbf{r},\mathbf{r}',\omega)$ is not literally well-defined at individual frequencies. Thus, at least on the level of final physical results, the effect of a discrete zero eigenvalue of the operator associated with $\boldsymbol{\sigma}(\mathbf{r},\mathbf{r}',\omega)$ cannot be significantly different from that of a very small but non-zero eigenvalue so that any method of regularizing $\boldsymbol{\rho}(\mathbf{r},\mathbf{r}',\omega)$ that implements this idea can be expected to give the same final results. The problem should hence be irrelevant in practice. One may simply perform all calculations for a class of amplifying media for which there are no problems with $\boldsymbol{\rho}(\mathbf{r},\mathbf{r}',\omega)$. Results obtained in this way, if they are physically understandable, can then be expected to hold also without restriction.

Finally, let us address the problem of the unboundedness from below of the energy eigenvalue spectrum attributed to the Hamiltonian (2.155) in the case of linear amplification. Because of the lack of a lower bound, the system could evolve into states of lower and lower energy by the creation of quanta in a frequency range where $\operatorname{sgn}\sigma(\alpha,\omega)=-1$, in which case the theory would gradually become unrealistic. If the system described by the Hamiltonian in Eq. (2.155) is coupled to a second system (e.g., an atom), another aspect of the problem is that the second system might (but need not) become more and more excited, which of course also becomes unrealistic at some stage. However, such catastrophes could only occur if the theory were used beyond its range of validity. In fact, they would indicate nothing but the breakdown of the concept of linear amplification. As long as the concept of linear amplification applies, the unboundedness from below of the energy eigenvalue spectrum may be regarded as being a purely formal drawback rather than a real one. Needless to say that this unboundedness prevents one from constructing the canonical density operator—the system cannot thermalize. Therefore, it should also be clear that electromagnetic-field correlation functions and fluctuation–dissipation relations in the familiar form known for absorbing media are not applicable to amplifying media, not even at zero temperature, i.e., to the vacuum state (see the pertinent critical remarks on Ref. [50] made in Ref. [R11] in this context). Nevertheless, using the results of this section, all desired correlation functions can be calculated straightforwardly for any well-defined quantum state (in particular, for the vacuum state).

Concluding our discussion of amplifying media and the whole chapter, we may say that the general quantization scheme is universally applicable to linearly responding absorbing media, and extends in a natural way also to media that are (linearly) amplifying. For the latter, the basic condition to apply the theory is the validity of the concept of linear amplification for the respective problem under study. It has turned out that when considering amplifying media more care and prudence is needed than in the case of robust equilibrium media, which only give rise to absorption. For a deeper understanding of the quantization scheme when it is applied to amplifying media, it would certainly be advantageous to have available also a more microscopic model of the quantized electromagnetic field interacting with linear media that also allow for amplification, especially an analog of the well-known Huttner-Barnett-type harmonic-oscillator models that are frequently used to study the quantized field in absorbing media [51–53]. Such a model should include a reservoir as in the case of absorbing media, but presumably also a second, 'inverted' reservoir capable of being prepared in a (formal) negative-temperature state. To our knowledge, Huttner-Barnett-type models that aim to incorporate amplification have unfortunately not yet been developed.

Chapter 3

Lorentz-Force Approach to Dispersion Forces

3.1 Introductory Remarks

Having developed the general quantization scheme in Chap. 2, we are now in a position to apply it to the problem of dispersion forces, i.e., forces mediated by the fluctuating vacuum. The remaining part of this work is devoted to this topic, on which (by application of the quantization scheme) we are adopting a macroscopic perspective. As mentioned already in Chap. 1, calculations of dispersion forces have often been based on expressions for the energy or the momentum flow (stress) attributed to the quantized electromagnetic field in media whose validity is debatable already at the classical level. We will see that such problems are avoided if the dispersion force is defined more directly, on the basis of the ground-state Lorentz force (density) in media. As a further motivation, let us first recapitulate a few facts from the theory of CP forces acting on single ground-state atoms in the presence of macroscopic bodies. The macroscopic bodies considered in the following are assumed to be absorbing ones; amplifying media are not considered.

The CP force acting on a ground-state atom in the vicinity of (linearly responding, absorbing) macroscopic bodies can be regarded as being a conservative force. Hence, it can be given by the negative gradient of a potential, which in the leading order of perturbation theory reads

$$\mathbf{F}^{(\mathrm{at})}(\mathbf{r}) = -\frac{\hbar\mu_0}{2\pi}\int_0^\infty \mathrm{d}\xi\,\xi^2\alpha(i\xi)\boldsymbol{\nabla}\mathrm{Tr}\,\boldsymbol{G}^{(\mathrm{S})}(\mathbf{r},\mathbf{r},i\xi), \tag{3.1}$$

where $\mathbf{r}$ is the position of the atom, $\alpha(i\xi)$ is its polarizability, and

$\boldsymbol{G}^{(\mathrm{S})}(\mathbf{r},\mathbf{r}',i\xi)$ is the scattering part of the Green tensor $\boldsymbol{G}(\mathbf{r},\mathbf{r}',i\xi)$ of the macroscopic Maxwell equations that take into account the bodies (but not the atom), evaluated at imaginary frequency $\omega = i\xi$. The potential (3.1) has been derived in the literature by various methods (see, e.g., Refs. [54–58]). In particular, in Ref. [58] it has been derived under the assumption that the bodies taken into account in the Green tensor $\boldsymbol{G}(\mathbf{r},\mathbf{r}',\omega)$ are magnetodielectric ones characterized by a local dielectric susceptibility $\varepsilon(\mathbf{r},\omega)-1$ and local (para-)magnetic susceptibility $1-\kappa(\mathbf{r},\omega)$ $[\kappa(\mathbf{r},\omega)=\mu^{-1}(\mathbf{r},\omega)]$. For such media, the Green tensor $\boldsymbol{G}(\mathbf{r},\mathbf{r}',\omega)$ obeys the equation

$$\boldsymbol{\nabla}\times\kappa(\mathbf{r},\omega)\boldsymbol{\nabla}\times\boldsymbol{G}(\mathbf{r},\mathbf{r}',\omega)-\frac{\omega^2}{c^2}\varepsilon(\mathbf{r},\omega)\boldsymbol{G}(\mathbf{r},\mathbf{r}',\omega)=\boldsymbol{I}\delta(\mathbf{r}-\mathbf{r}'), \tag{3.2}$$

together with the boundary condition at infinity. The scattering part of the Green tensor figuring in (3.1) contains all the necessary information about the configuration of the locally responding magnetodielectric bodies and is well-defined in the limit of coincident spatial arguments. The (translationally invariant) bulk part of the Green tensor on the other hand, which would diverge in the coincidence limit, is not needed in Eq. (3.1), because it cannot contribute to the force. Equation (3.1) strictly applies to isolated atoms, but not to medium atoms nor to guest atoms in a substrate medium. The atomic ground-state polarizability $\alpha(\omega)$ in leading-order perturbation theory,

$$\alpha(\omega)\sim\sum_k\frac{\Omega_k^2}{\omega_k^2-\omega^2}\,, \tag{3.3}$$

features poles on the real frequency axis due to the neglect of level broadening. If necessary, the correct response function properties [35] may be restored by means of an appropriate limit prescription, viz.

$$\alpha(\omega)\sim\lim_{\gamma\to0+}\sum_k\frac{\Omega_k^2}{\omega_k^2-\omega^2-i\gamma\omega}\,. \tag{3.4}$$

In order to apply Eq. (3.1) to a (locally responding) dielectric body of volume V_{M}, let us consider instead of a single atom a collection of atoms that are (strictly) contained inside a space region of volume V_{M}, and let us add up the individual forces as given by Eq. (3.1). Since the mutual interaction of the atoms is completely ignored in this way, it is clear that this method gives only the lowest-order approximation to the total force. If the number density of the atoms (defined on a suitably chosen macroscopic length scale) is denoted by $\eta(\mathbf{r})$, the total force in this approximation reads

$$\mathbf{F} = -\frac{\hbar\mu_0}{2\pi}\int_{V_\mathrm{M}} \mathrm{d}^3r \int_0^\infty \mathrm{d}\xi\, \xi^2 \eta(\mathbf{r})\alpha(i\xi)\boldsymbol{\nabla}\mathrm{Tr}\, \boldsymbol{G}^{(\mathrm{S})}(\mathbf{r},\mathbf{r},i\xi). \tag{3.5}$$

Since the validity of Eq. (3.5) obviously requires sufficiently weakly polarizable atoms and/or a sufficiently low number density of atoms, the collection of atoms can be viewed as dielectric matter of volume V_M and small susceptibility

$$\chi_\mathrm{M}(\mathbf{r},i\xi) = \eta(\mathbf{r})\alpha(i\xi)/\varepsilon_0, \tag{3.6}$$

which implies that the permittivity of the overall system has slightly been changed by $\delta\varepsilon(\mathbf{r},i\xi) = \chi_\mathrm{M}(\mathbf{r},i\xi)$. In particular, applying Eq. (3.5) to a dielectric (micro-)object whose number density of atoms is constant over the (small) volume V_M, we obtain the force

$$\mathbf{F} = -V_\mathrm{M}\eta\,\frac{\hbar\mu_0}{2\pi}\int_0^\infty \mathrm{d}\xi\, \xi^2 \alpha(i\xi)\boldsymbol{\nabla}\mathrm{Tr}\, \boldsymbol{G}^{(\mathrm{S})}(\mathbf{r},\mathbf{r},i\xi). \tag{3.7}$$

Note that Eqs. (3.5) and (3.7) correspond to the result that is found by pairwise summation of vdW interactions [10, 59] and represent, with respect to the weakly polarizable object, nothing but the leading term of the exact sum over all the many-atom vdW forces [19–21]. Equations of the type (3.7) have widely been used to study dispersive forces on weakly polarizable matter (see, e.g., Refs. [5, 11, 13] and [R4]).

As known from Sec. 2.4, a locally responding (magneto-)dielectric medium may be described by a conductivity tensor $\boldsymbol{Q}(\mathbf{r},\mathbf{r}',\omega)$ of the specific quasilocal form (2.142), and Eq. (3.2) is nothing but the specialization of Eq. (2.18) to this case. Moreover, since from Eqs. (3.1) and (3.5) one cannot see what class of linear media has been allowed in the construction of the Green tensor $\boldsymbol{G}(\mathbf{r},\mathbf{r}',\omega)$, we may ask if they will retain their validity also if general linear media (rather than locally responding magnetodielectrics) are considered. Furthermore, it is clear that application of Eq. (3.5) to dielectric bodies (including micro-objects) that are dense and/or consist of strongly polarizable atoms becomes questionable. Hence, the problem of a generalization of Eq. (3.5) to arbitrary dielectric bodies (rather than only weakly dielectric ones) or parts of them arises. Instead of determining the exact Casimir force on a body by summing up, in one or another way, the (many-atom) dispersion forces on microscopic levels, we shall approach the problem from the opposite side and obtain a formulation of the Casimir force that generalizes Eq. (3.5) and enables one to analyze the Casimir force into constituent CP and/or vdW forces, thereby making most transparent the common character of the dispersion forces. We begin by preparing a firm ground for the definition of the dispersion force density in media.

3.2 Dispersion Forces as Lorentz Forces

As known, the classical Lorentz force density

$$\mathbf{f} = \rho\mathbf{E} + \mathbf{j}\times\mathbf{B} \tag{3.8}$$

may be rewritten in the equivalent form

$$\mathbf{f} = \boldsymbol{\nabla}\cdot\boldsymbol{T} - \varepsilon_0\frac{\partial}{\partial t}(\mathbf{E}\times\mathbf{B}), \tag{3.9}$$

with $\boldsymbol{T}$ being Maxwell's stress tensor

$$\boldsymbol{T} = \varepsilon_0\mathbf{E}\mathbf{E} + \mu_0^{-1}\mathbf{B}\mathbf{B} - \tfrac{1}{2}(\varepsilon_0 E^2 + \mu_0^{-1}B^2)\boldsymbol{I}, \tag{3.10}$$

which is symmetric, $\boldsymbol{T} = \boldsymbol{T}^{\mathsf{T}}$. If all the charges and currents of the system under consideration have been included in ρ and $\mathbf{j}$, respectively, Eqs. (3.8) and (3.9) [with Eq. (3.10)] are universally valid, regardless if the charges and currents are viewed as forming a medium, i.e., regardless if any constitutive relation(s) have been assumed. Note that essentially the same position has been taken up in Ref. [60] in the (re)analysis of classical force experiments measuring the electromagnetic force that acts on dielectric [61–65] or magnetodielectric [66] (see also Refs. [67,68]) disks exposed to crossed electric and magnetic fields. The integral of the Lorentz force density $\mathbf{f}$ over some space region V gives the (in general time-dependent) total electromagnetic force $\mathbf{F}$ acting on the matter inside it,

$$\mathbf{F} = \int_V \mathrm{d}^3r\,\mathbf{f}, \tag{3.11}$$

which, from Eq. (3.9), may be equivalently written as

$$\mathbf{F} = \int_{\partial V}\mathrm{d}\mathbf{a}\cdot\boldsymbol{T} - \varepsilon_0\frac{\mathrm{d}}{\mathrm{d}t}\int_V \mathrm{d}^3r\,\mathbf{E}\times\mathbf{B}, \tag{3.12}$$

regardless if the space region V is occupied by a macroscopic body or not. In particular, under stationary conditions the volume integral on the right-hand side of Eq. (3.12) does not contribute to the (slowly varying part of the) total force, so that the force may be found by integrating the stress tensor over a closed surface. In so doing, a constant term in the stress tensor can be omitted as it obviously does not contribute.

The idea to regard [according to Eqs. (3.8)–(3.11)] the Lorentz force acting on the totality of charges and currents belonging (on the chosen scale) to a system under consideration as the fundamental quantity is neither new [60, 69–71] nor particularly hard to agree with, but the use of Minkowski's stress tensor or related quantities has nevertheless been common in the calculation of electromagnetic forces. Generally, the idea behind using Minkowski's stress tensor is to include further ('mechanical') force contributions besides the electromagnetic ones in the momentum balance. However, as shown in Refs. [R5] and [R6], Minkowski's stress tensor is not satisfactory in this regard and may lead to contradictory results in general. Focusing on genuinely electromagnetic forces, we therefore adopt the Lorentz force, and with it Maxwell's stress tensor (3.10), as the suitable basis for the calculation of dispersion forces. In Ref. [R3] we have shown that this point of view is also consistent with microscopic harmonic-oscillator models of dispersing and absorbing (locally responding dielectric) matter, which are generally accepted and frequently used in the literature.

The above classical considerations hold similarly in quantum theory. Assuming that all the charges and currents present in space may be attributed to a linear medium with the conductivity tensor $\boldsymbol{Q}(\mathbf{r},\mathbf{r}',\omega)$, the medium-assisted electric field operator $\hat{\mathbf{E}}(\mathbf{r})$ and the induction field operator $\hat{\mathbf{B}}(\mathbf{r})$ are given by Eqs. (2.20) and (2.21) with Eqs. (2.22) and (2.23), respectively, where $\underline{\hat{\mathbf{j}}}_{\mathrm{N}}(\mathbf{r},\omega)$ obeys the commutation relation Eq. (2.25). The charge and current densities that are subject to the Lorentz force are given by

$$\hat{\rho}(\mathbf{r}) = \int_0^\infty \mathrm{d}\omega\, \underline{\hat{\rho}}(\mathbf{r},\omega) + \mathrm{H.c.}, \tag{3.13}$$

$$\hat{\mathbf{j}}(\mathbf{r}) = \int_0^\infty \mathrm{d}\omega\, \underline{\hat{\mathbf{j}}}(\mathbf{r},\omega) + \mathrm{H.c.}, \tag{3.14}$$

with

$$\underline{\hat{\rho}}(\mathbf{r},\omega) = (i\omega)^{-1}\boldsymbol{\nabla}\cdot\underline{\hat{\mathbf{j}}}(\mathbf{r},\omega) \tag{3.15}$$

and [cf. Eqs. (2.8) and (2.17)]

$$\underline{\hat{\mathbf{j}}}(\mathbf{r},\omega) = i\mu_0\omega\int \mathrm{d}^3r' \int \mathrm{d}^3s\, \boldsymbol{Q}(\mathbf{r},\mathbf{r}',\omega)\cdot\boldsymbol{G}(\mathbf{r}',\mathbf{s},\omega)\cdot\underline{\hat{\mathbf{j}}}_{\mathrm{N}}(\mathbf{s},\omega) + \underline{\hat{\mathbf{j}}}_{\mathrm{N}}(\mathbf{r},\omega), \tag{3.16}$$

respectively. Recall that $\hat{\underline{\mathbf{j}}}_{\mathrm{N}}(\mathbf{r},\omega)$ should be thought of as being expressed, according to Eq. (2.32), in terms of the bosonic dynamical variables $\hat{\mathbf{f}}(\mathbf{r},\omega)$ used to define the state space [or alternatively, according to Eq. (2.53), in terms of the $\hat{g}(\alpha,\omega)$]. One can prove (App. A.6) that

$$\big[\hat{\rho}(\mathbf{r}),\hat{\mathbf{E}}(\mathbf{r}')\big]=0, \tag{3.17}$$

$$\big[\hat{\rho}(\mathbf{r}),\hat{\mathbf{B}}(\mathbf{r}')\big]=0, \tag{3.18}$$

$$\big[\hat{\mathbf{j}}(\mathbf{r}),\hat{\mathbf{B}}(\mathbf{r}')\big]=0, \tag{3.19}$$

and

$$\big[\hat{\mathbf{j}}(\mathbf{r}),\hat{\mathbf{E}}(\mathbf{r}')\big]=\frac{i\hbar}{\varepsilon_0}\,\mathrm{Im}\,\boldsymbol{Q}^{(-1)}(\mathbf{r},\mathbf{r}'), \tag{3.20}$$

where $\boldsymbol{Q}^{(-1)}(\mathbf{r},\mathbf{r}')$ is defined via the leading asymptotic behavior of $\boldsymbol{Q}(\mathbf{r},\mathbf{r}',\omega)$ as ω approaches infinity along any direction in the upper ω half-plane, according to

$$\boldsymbol{Q}(\mathbf{r},\mathbf{r}',\omega)\simeq\boldsymbol{Q}^{(-1)}(\mathbf{r},\mathbf{r}')/\omega. \tag{3.21}$$

The coefficient $\boldsymbol{Q}^{(-1)}(\mathbf{r},\mathbf{r}')$ is purely imaginary since $\boldsymbol{Q}(\mathbf{r},\mathbf{r}',\omega)$ is purely real on the imaginary frequency axis. In the special case where $\boldsymbol{Q}(\mathbf{r},\mathbf{r}',\omega)$ corresponds to a locally responding (isotopic) magnetodielectric medium [see Eqs.(2.142), (2.143)], Eqs. (3.15) and (3.16) may be written in the form

$$\hat{\underline{\rho}}(\mathbf{r},\omega)=-\varepsilon_0\boldsymbol{\nabla}\cdot\{[\varepsilon(\mathbf{r},\omega)-1]\hat{\underline{\mathbf{E}}}(\mathbf{r},\omega)\}+(i\omega)^{-1}\boldsymbol{\nabla}\cdot\hat{\underline{\mathbf{j}}}_{\mathrm{N}}(\mathbf{r},\omega) \tag{3.22}$$

and

$$\begin{aligned}\hat{\underline{\mathbf{j}}}(\mathbf{r},\omega)=&-i\omega\varepsilon_0[\varepsilon(\mathbf{r},\omega)-1]\hat{\underline{\mathbf{E}}}(\mathbf{r},\omega)\\&+\boldsymbol{\nabla}\times\{\kappa_0[1-\kappa(\mathbf{r},\omega)]\hat{\underline{\mathbf{B}}}(\mathbf{r},\omega)\}+\hat{\underline{\mathbf{j}}}_{\mathrm{N}}(\mathbf{r},\omega),\end{aligned} \tag{3.23}$$

respectively. For such a medium, one finds that

$$\boldsymbol{Q}^{(-1)}(\mathbf{r},\mathbf{r}')=i\varepsilon_0\Omega_\varepsilon^2(\mathbf{r})\boldsymbol{I}\delta(\mathbf{r}-\mathbf{r}'), \tag{3.24}$$

where the position-dependent (and real) plasma frequency $\Omega_\varepsilon(\mathbf{r})$ is defined by the asymptotic behavior of the permittivity in the upper ω half-plane according to $\varepsilon(\mathbf{r},\omega)\simeq 1-\Omega_\varepsilon^2(\mathbf{r})/\omega^2$. Note that the magnetic permeability $\mu(\mathbf{r},\omega)=\kappa^{-1}(\mathbf{r},\omega)$ does not contribute to Eq. (3.24). Also note that the plasma frequency $\Omega_\varepsilon(\mathbf{r})$, and more generally the quantity $\boldsymbol{Q}^{(-1)}(\mathbf{r},\mathbf{r}')$, can generally be assumed to exist as a finite quantity, on the grounds that any medium ultimately behaves like a gas (plasma) of free charged particles [31] at sufficiently high frequencies.

The commutation relations (3.17)–(3.20) show that $\hat{\rho}(\mathbf{r})$ and $\hat{\mathbf{j}}(\mathbf{r})$ really represent matter quantities. However, Eq. (3.20) may appear surprising at first glance as it depends on the specific medium. To understand this, we note that in the special case of Eq. (3.24) it follows from Eq. (3.20) that

$$\big[\hat{\mathbf{j}}(\mathbf{r}), \hat{\mathbf{E}}^{\perp}(\mathbf{r}')\big] = i\hbar\,\Omega_{\varepsilon}^{2}(\mathbf{r})\boldsymbol{\Delta}_{\perp}(\mathbf{r}-\mathbf{r}'). \tag{3.25}$$

The latter equation may be confirmed explicitly when—on the basis of a microscopic description—the current density is explicitly specified in terms of particle velocities. Carrying out the derivation in the minimal coupling scheme (see App. A.6), it is seen that the particle-related microscopic current density fails to commute with (the microscopic) $\hat{\mathbf{E}}^{\perp}(\mathbf{r})$ precisely since the (Coulomb gauged) vector potential modifies the canonical particle momenta as compared to the decoupled situation, which on macroscopic scales leads to Eq. (3.25). Similarly, Eq. (3.20) reflects that the matter described by $\hat{\rho}(\mathbf{r})$ and $\hat{\mathbf{j}}(\mathbf{r})$ is not decoupled from the field. In order to introduce the coupling to the (microscopic) electromagnetic field, the pertinent microscopic matter operators have already been modified, which on the level of the macroscopic description eventually manifests itself in the commutator Eq. (3.20).

In order to evaluate the quantum-mechanical expectation value of the operator Lorentz force, an appropriate density operator should be assigned to the overall system, where in the context of dispersion forces we have to consider the ground state (vacuum). It is also customary, on the other hand, to to allow for the system being at finite temperature. The fluctuation-induced forces at finite temperature may be viewed as being dispersion forces in a wider sense, but in contrast to the ground-state forces, they are not entirely quantum-mechanical (as thermal fluctuations—unlike vacuum fluctuations—have a classical counterpart). Clearly, if the field-matter system is in a number state [defined with respect to the number (density) operators $\hat{\mathbf{f}}^{\dagger}(\mathbf{r},\omega)\cdot\hat{\mathbf{f}}(\mathbf{r},\omega)$] such as the ground state, or an incoherent mixture of them such as a thermal state, then all one-time averages are time-independent. Recalling the bosonic character of the fundamental fields $\hat{\mathbf{f}}(\mathbf{r},\omega)$ and $\hat{\mathbf{f}}^{\dagger}(\mathbf{r},\omega)$ [cf. Eq. (2.31)] and assuming them to be excited thermally, i.e.,

$$\hat{\rho} = e^{-\hat{H}/(k_{\mathrm{B}}T)}/[\mathrm{Tr}\, e^{-\hat{H}/(k_{\mathrm{B}}T)}] \tag{3.26}$$

with $\hat{H}$ from Eq. (2.30), one can show that

$$\left\langle \hat{\mathbf{f}}(\mathbf{r},\omega)\hat{\mathbf{f}}^\dagger(\mathbf{r}',\omega')\right\rangle = \tfrac{1}{2}\left[\coth\!\left(\frac{\hbar\omega}{2k_\mathrm{B}T}\right)+1\right]\delta(\omega-\omega')\boldsymbol{I}\delta(\mathbf{r}-\mathbf{r}'), \tag{3.27}$$

$$\left\langle \hat{\mathbf{f}}^\dagger(\mathbf{r},\omega)\hat{\mathbf{f}}(\mathbf{r}',\omega')\right\rangle = \tfrac{1}{2}\left[\coth\!\left(\frac{\hbar\omega}{2k_\mathrm{B}T}\right)-1\right]\delta(\omega-\omega')\boldsymbol{I}\delta(\mathbf{r}-\mathbf{r}'), \tag{3.28}$$

and

$$\left\langle \hat{\mathbf{f}}(\mathbf{r},\omega)\hat{\mathbf{f}}(\mathbf{r}',\omega')\right\rangle = 0. \tag{3.29}$$

Making use of Eqs.(2.32) and (2.33), the correlation functions (3.27)–(3.29) are easily seen to imply the correlation functions [cf. Eq. (2.10)]

$$\left\langle \underline{\hat{\mathbf{j}}}_\mathrm{N}(\mathbf{r},\omega)\underline{\hat{\mathbf{j}}}_\mathrm{N}^\dagger(\mathbf{r}',\omega')\right\rangle = \frac{\hbar\omega}{2\pi}\,\delta(\omega-\omega')\left[\coth\!\left(\frac{\hbar\omega}{2k_\mathrm{B}T}\right)+1\right]\boldsymbol{\sigma}(\mathbf{r},\mathbf{r}',\omega), \tag{3.30}$$

$$\left\langle \underline{\hat{\mathbf{j}}}_\mathrm{N}^\dagger(\mathbf{r},\omega)\underline{\hat{\mathbf{j}}}_\mathrm{N}(\mathbf{r}',\omega')\right\rangle = \frac{\hbar\omega}{2\pi}\,\delta(\omega-\omega')\left[\coth\!\left(\frac{\hbar\omega}{2k_\mathrm{B}T}\right)-1\right]\boldsymbol{\sigma}(\mathbf{r},\mathbf{r}',\omega), \tag{3.31}$$

and

$$\langle \underline{\hat{\mathbf{j}}}_\mathrm{N}(\mathbf{r},\omega)\underline{\hat{\mathbf{j}}}_\mathrm{N}(\mathbf{r}',\omega')\rangle = 0. \tag{3.32}$$

As the zero-temperature limit of Eq. (3.26) is the ground-state projector, the corresponding ground-state correlation functions may be obtained by performing this limit in Eqs. (3.27), (3.28), (3.30) and (3.31), effectively replacing the hyperbolic cotangent with unity.

In order to prove Eqs. (3.27)–(3.29), one may discretize the continuum of Bose variables and perform the calculations, e.g., in the occupation-number representation (see Ref. [R1]). For a more elegant derivation (without discretization) one may resort to the normal-ordering formula (normal-ordering notation $:\cdots:$)

$$\begin{aligned}&\exp\left[\int_0^\infty \mathrm{d}\omega\int \mathrm{d}^3r\,\lambda(\mathbf{r},\omega)\hat{\mathbf{f}}^\dagger(\mathbf{r},\omega)\cdot\hat{\mathbf{f}}(\mathbf{r},\omega)\right]\\ &\qquad =:\exp\left\{\int_0^\infty \mathrm{d}\omega\int \mathrm{d}^3r\,[e^{\lambda(\mathbf{r},\omega)}-1]\hat{\mathbf{f}}^\dagger(\mathbf{r},\omega)\cdot\hat{\mathbf{f}}(\mathbf{r},\omega)\right\}:\,.\end{aligned} \tag{3.33}$$

The derivation of Eq. (3.33) can be based on the (functional) differential equation $\delta\hat{F}^{(\mathrm{N})}/\delta\lambda(\mathbf{r},\omega) = \hat{F}^{(\mathrm{N})}\hat{\mathbf{f}}^\dagger(\mathbf{r},\omega)\cdot\hat{\mathbf{f}}(\mathbf{r},\omega)$ where $\hat{F}^{(\mathrm{N})} = \hat{F}^{(\mathrm{N})}[\lambda(\mathbf{r},\omega)]$ is defined as the equivalent normally-ordered functional form of the operator on the left-hand side of Eq. (3.33). Using Eq. (2.31), one can see that the differential equation may be rewritten as $\delta\hat{F}^{(\mathrm{N})}/\delta\lambda(\mathbf{r},\omega) = \exp[\lambda(\mathbf{r},\omega)]\hat{\mathbf{f}}^\dagger(\mathbf{r},\omega)\cdot$

$\hat{F}^{(\mathrm{N})}\hat{\mathbf{f}}(\mathbf{r},\omega) =: \exp[\lambda(\mathbf{r},\omega)]\hat{\mathbf{f}}^\dagger(\mathbf{r},\omega)\hat{F}^{(\mathrm{N})}\hat{\mathbf{f}}(\mathbf{r},\omega) :$, where the last step is valid because $\hat{F}^{(\mathrm{N})}$ is in normal order by assumption. Since ordinary (commutative) calculus is applicable between the normal ordering symbols, the equation may now be solved straightforwardly; taking into account that $\hat{F}^{(\mathrm{N})}[\lambda(\mathbf{r},\omega)]$ is the unit operator if $\lambda(\mathbf{r},\omega)$ is the null function, one arrives at Eq. (3.33). Rewriting the density operator (3.26) in normal order by means of Eq. (3.33) $[\lambda(\mathbf{r},\omega) \mapsto -\hbar\omega/(k_\mathrm{B}T)]$, one may read off the phase-space function(al) appropriate for the calculation of averages of anti-normally ordered operators [Q-function(al)] (see, e.g., Refs. [42,72]), and confirm Eqs. (3.27) [and thereby also Eq. (3.28)] and Eq. (3.29) by performing a simple phase-space integral. From the derivation it is evident that Eqs. (3.27)–(3.29) would remain valid in the same form even if the global temperature T were replaced with a temperature field $T(\mathbf{r})$.

3.2.1 Stress-Tensor Formulation

Now we can calculate the expectation value of the Lorentz force operator [which is Hermitean—recall Eqs. (3.17) and (3.19)],

$$\mathbf{F} = \int_V \mathrm{d}^3 r \, \langle \hat{\rho}\hat{\mathbf{E}} + \hat{\mathbf{j}} \times \hat{\mathbf{B}} \rangle, \tag{3.34}$$

where $\hat{\mathbf{E}}$ and $\hat{\mathbf{B}}$, respectively, are defined by Eqs. (2.20) and (2.21) together with Eqs. (2.22) and (2.23), and $\hat{\rho}$ and $\hat{\mathbf{j}}$, respectively, are defined by Eqs. (3.13) and (3.14) together with Eqs. (3.22) and (3.23). Following the line suggested by classical electrodynamics, paying proper attention to operator symmetrization as well as regularization, and taking into account that the time derivative in the (quantum-mechanical version of) Eq. (3.12) does not contribute to the force, one can show (see Ref. [R3]) that the force may be calculated as a surface integral $\mathbf{F} = \int_{\partial V} \mathrm{d}\mathbf{a} \cdot \boldsymbol{T}$ over the (symmetric, time-independent) stress tensor, which is obtained [in agreement with the classical Eq. (3.10)] from the quantum-mechanical expectation value

$$\begin{aligned} \boldsymbol{T}(\mathbf{r},\mathbf{r}') = {} & \varepsilon_0 \langle \hat{\mathbf{E}}(\mathbf{r})\hat{\mathbf{E}}(\mathbf{r}') \rangle + \mu_0^{-1} \langle \hat{\mathbf{B}}(\mathbf{r})\hat{\mathbf{B}}(\mathbf{r}') \rangle \\ & - \tfrac{1}{2}\boldsymbol{I}\left[\varepsilon_0 \langle \hat{\mathbf{E}}(\mathbf{r}) \cdot \hat{\mathbf{E}}(\mathbf{r}') \rangle + \mu_0^{-1} \langle \hat{\mathbf{B}}(\mathbf{r}) \cdot \hat{\mathbf{B}}(\mathbf{r}') \rangle\right] \end{aligned} \tag{3.35}$$

in the limit $\mathbf{r}' \to \mathbf{r}$, where divergent bulk contributions (corresponding to unphysical self-forces) are to be removed before taking the limit. Note that in the calculation of the surface integral over the stress tensor the 'outer' values of the integrand should in general be used. Using Eqs. (2.20) and (2.21) together with Eqs. (2.22) and (2.23) and (3.30)–(3.32) and employing (2.19) and the reciprocity property of the Green tensor, one can calculate the thermal-equilibrium correlation functions of the electric field and the induction field as

$$\langle \hat{\mathbf{E}}(\mathbf{r})\hat{\mathbf{E}}(\mathbf{r}')\rangle = \frac{\hbar\mu_0}{\pi}\int_0^\infty \mathrm{d}\omega\,\omega^2 \coth\left(\frac{\hbar\omega}{2k_\mathrm{B}T}\right)\mathrm{Im}\,\boldsymbol{G}(\mathbf{r},\mathbf{r}',\omega), \tag{3.36}$$

$$\langle \hat{\mathbf{B}}(\mathbf{r})\hat{\mathbf{B}}(\mathbf{r}')\rangle = -\frac{\hbar\mu_0}{\pi}\int_0^\infty \mathrm{d}\omega\,\coth\left(\frac{\hbar\omega}{2k_\mathrm{B}T}\right)\boldsymbol{\nabla}\times\mathrm{Im}\,\boldsymbol{G}(\mathbf{r},\mathbf{r}',\omega)\times\overleftarrow{\boldsymbol{\nabla}}'. \tag{3.37}$$

The correlation functions (3.36) and (3.37) inherit the reciprocity property of the Green tensor. Inserting Eqs. (3.36) and (3.37) into Eq. (3.35) finally yields the stress tensor of the dispersion force as

$$\boldsymbol{T}(\mathbf{r},\mathbf{r}) = \lim_{\mathbf{r}'\to\mathbf{r}}\left[\boldsymbol{\theta}(\mathbf{r},\mathbf{r}') - \tfrac{1}{2}\boldsymbol{I}\,\mathrm{Tr}\,\boldsymbol{\theta}(\mathbf{r},\mathbf{r}')\right], \tag{3.38}$$

where

$$\boldsymbol{\theta}(\mathbf{r},\mathbf{r}') = \frac{\hbar}{\pi}\int_0^\infty \mathrm{d}\omega\,\coth\left(\frac{\hbar\omega}{2k_\mathrm{B}T}\right) \times\left[\frac{\omega^2}{c^2}\,\mathrm{Im}\,\boldsymbol{G}(\mathbf{r},\mathbf{r}',\omega) - \boldsymbol{\nabla}\times\mathrm{Im}\,\boldsymbol{G}(\mathbf{r},\mathbf{r}',\omega)\times\overleftarrow{\boldsymbol{\nabla}}'\right]. \tag{3.39}$$

As expected, the conductivity tensor $\boldsymbol{Q}(\mathbf{r},\mathbf{r}',\omega)$ does not appear explicitly in Eqs. (3.38), (3.39), but only via the Green tensor $\boldsymbol{G}(\mathbf{r},\mathbf{r}',\omega)$. As mentioned, by performing the limit $T\to 0$ (i.e., by replacing the hyperbolic cotangent with unity) one can find the corresponding ground-state result.

Having removed divergent bulk contributions, we may take the imaginary part of the whole integral instead of the integrand in Eq. (3.39) and rotate (in view of the analytical properties of the integrand), the integration contour toward the imaginary frequency axis, on which the Green tensor is real [$\boldsymbol{G}^*(\mathbf{r},\mathbf{r}',\omega) = \boldsymbol{G}(\mathbf{r},\mathbf{r}',-\omega^*)$.] In the zero-temperature limit, the result is simply

$$\boldsymbol{\theta}(\mathbf{r},\mathbf{r}') = -\frac{\hbar}{\pi}\int_0^\infty \mathrm{d}\xi\,\left[\frac{\xi^2}{c^2}\,\boldsymbol{G}(\mathbf{r},\mathbf{r}',i\xi) + \boldsymbol{\nabla}\times\boldsymbol{G}(\mathbf{r},\mathbf{r}',i\xi)\times\overleftarrow{\boldsymbol{\nabla}}'\right]. \tag{3.40}$$

For non-zero temperatures, the (first-order) poles of the hyperbolic cotangent at the imaginary (Matsubara) frequencies $\omega_m = i\xi_m = 2im\pi k_\mathrm{B}T/\hbar$ (m, integer),

$$\coth\left(\frac{\hbar\omega}{2k_\mathrm{B}T}\right) = \frac{2k_\mathrm{B}T}{\hbar}\sum_{m=0}^{\infty}\left(1-\frac{\delta_{m0}}{2}\right)\left(\frac{1}{\omega-\omega_m}+\frac{1}{\omega+\omega_m}\right), \tag{3.41}$$

must be taken into account. Instead of Eq. (3.40), one then finds

$$\boldsymbol{\theta}(\mathbf{r},\mathbf{r}') = -2k_{\mathrm{B}}T \sum_{m=0}^{\infty} \left(1 - \frac{\delta_{m0}}{2}\right) \times \left[\frac{\xi^2}{c^2}\,\boldsymbol{G}(\mathbf{r},\mathbf{r}',i\xi) + \boldsymbol{\nabla}\times\boldsymbol{G}(\mathbf{r},\mathbf{r}',i\xi)\times\overleftarrow{\boldsymbol{\nabla}}'\right]_{\xi=\xi_m}. \quad (3.42)$$

[In view of Eq. (3.40), this amounts to the well-known formal recipe $\int_0^\infty \mathrm{d}\xi \cdots \mapsto (2\pi k_{\mathrm{B}}T)\hbar^{-1}\sum_{m=0}^{\infty}(1-\delta_{m0}/2)\cdots$.] The derivation of Eq. (3.42) from Eq. (3.39) requires that the expression in the square bracket in Eq. (3.42) is non-singular at zero frequency. This is in fact the case, provided the (model) conductivity tensor used in the construction of $\boldsymbol{G}(\mathbf{r},\mathbf{r}',\omega)$ is in line with the consistency conditions expressed by Eqs. (A.9) and (A.10). The inclusion of finite temperature into the theory is therefore straightforward, as (on the level of two-point correlation functions) it merely amounts to the proper incorporation of the thermal factor $\coth[\hbar\omega/(2k_{\mathrm{B}}T)]$. In the following, we will thus restrict attention to the zero-temperature limit, where the system can be assumed to be in its ground state.

Before we proceed, let us briefly comment on locally responding media described by a Drude-type permittivity, which exhibits a (first-order) pole at zero frequency. Such media correspond to a conductivity tensor whose static limit is non-zero but violates the transversality condition (A.10). It is therefore not surprising that the zero-frequency ($m=0$) contribution to the series in Eq. (3.42) can be problematic for such media. The latter problem has attracted much attention in the literature and has given rise to controversial debates [73–76], which apparently have not led to a consensus. From App. A.2, we can conclude that the root of the problem is that Drude-type permittivities do not give rise to reasonable electrostatics and/or magnetostatics relations in the zero-frequency limit. The zero-frequency terms of formulas like Eq. (3.42) are thus not to be blamed for their failure in the case of Drude-model media—it is the Drude-model itself that fails, in the sense that it neglects spatial dispersion in a manner that becomes invalid in the zero-frequency limit.

3.2.2 Volume-Integral Formulation

In order to study the force (3.34) more directly, it may be useful to focus on the force density rather than the stress tensor. Let us hence calculate the (zero-temperature) dispersion force acting on a linearly responding body in some space region of volume V by direct evaluation of Eq. (3.34). We want to study—mostly in parallel—the two cases sketched in Figs. 3.1 and 3.2, namely (i) an isolated body (Fig. 3.1), and (ii) a body that is an inner part of some larger body (Fig. 3.2). In both cases, arbitrary linearly responding bodies are allowed to be present in the outer region V_B in the figures. Thus,

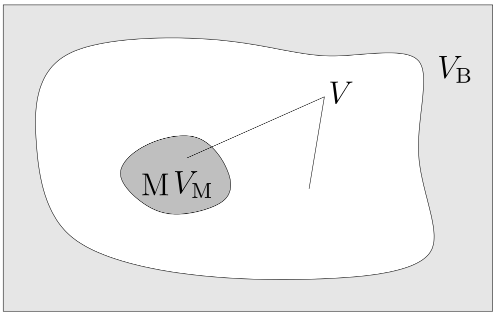

Figure 3.1: A body M of volume V_M inside an empty-space region of total volume V. There may be arbitrary bodies in the outer region V_B.

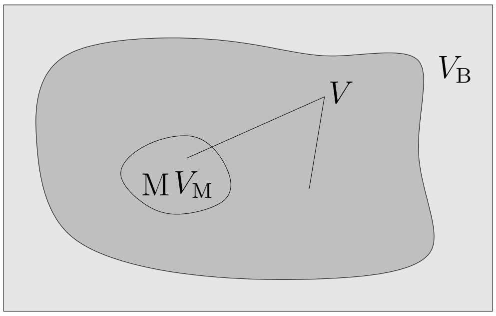

Figure 3.2: A body M of volume V_M that is an inner part of a larger body of volume V. There may be arbitrary bodies in the outer region V_B.

we have to express the density of the dispersion force that acts (in the zero-temperature limit) on the material in a chosen spatial region of volume V_M, i.e., the integrand of the volume integral

$$\mathbf{F} = \int_{V_\mathrm{M}} \mathrm{d}^3 r \int_0^\infty \mathrm{d}\omega \int_0^\infty \mathrm{d}\omega' \, \langle \underline{\hat{\rho}}(\mathbf{r},\omega) \underline{\hat{\mathbf{E}}}^\dagger(\mathbf{r}',\omega') + \underline{\hat{\mathbf{j}}}(\mathbf{r},\omega) \times \underline{\hat{\mathbf{B}}}^\dagger(\mathbf{r}',\omega') \rangle_{\mathbf{r}'\to\mathbf{r}}, \tag{3.43}$$

in terms of the Green tensor of the system. In performing the limit $\mathbf{r}' \to \mathbf{r}$ in Eq. (3.43), one must again omit unphysical self-force contributions. Using Eqs. (2.22), (2.23), and (2.18), one may show that $\underline{\hat{\rho}}(\mathbf{r},\omega)$ and $\underline{\hat{\mathbf{j}}}(\mathbf{r},\omega)$ as defined by Eqs. (3.15) and (3.16) can be written alternatively as

$$\underline{\hat{\rho}}(\mathbf{r},\omega) = \frac{i\omega}{c^2} \boldsymbol{\nabla} \cdot \int \mathrm{d}^3 r' \, \boldsymbol{G}(\mathbf{r},\mathbf{r}',\omega) \cdot \underline{\hat{\mathbf{j}}}_\mathrm{N}(\mathbf{r}',\omega), \tag{3.44}$$

$$\underline{\hat{\mathbf{j}}}(\mathbf{r},\omega) = \left(\boldsymbol{\nabla} \times \boldsymbol{\nabla} \times - \frac{\omega^2}{c^2} \right) \int \mathrm{d}^3 r' \, \boldsymbol{G}(\mathbf{r},\mathbf{r}',\omega) \cdot \underline{\hat{\mathbf{j}}}_\mathrm{N}(\mathbf{r}',\omega). \tag{3.45}$$

Taking into account (the zero-temperature versions of) Eqs.(3.30)–(3.32) and recalling the properties of the Green tensor, it follows from Eqs. (2.22), (2.23), (3.44) and (3.45) that

$$\langle\underline{\hat{\rho}}(\mathbf{r},\omega)\underline{\hat{\mathbf{E}}}^\dagger(\mathbf{r}',\omega')\rangle = \frac{\hbar}{\pi}\frac{\omega^2}{c^2}\,\delta(\omega-\omega')\boldsymbol{\nabla}\cdot\mathrm{Im}\,\boldsymbol{G}(\mathbf{r},\mathbf{r}',\omega) \tag{3.46}$$

and

$$\begin{aligned}&\langle\underline{\hat{\mathbf{j}}}(\mathbf{r},\omega)\underline{\hat{\mathbf{B}}}^\dagger(\mathbf{r}',\omega')\rangle\\ &\quad= -\frac{\hbar}{\pi}\,\delta(\omega-\omega')\left(\boldsymbol{\nabla}\times\boldsymbol{\nabla}\times-\frac{\omega^2}{c^2}\right)\mathrm{Im}\,\boldsymbol{G}(\mathbf{r},\mathbf{r}',\omega)\times\overleftarrow{\boldsymbol{\nabla}}'.\end{aligned} \tag{3.47}$$

In the following we denote by $\boldsymbol{G}_V(\mathbf{r},\mathbf{r}',\omega)$ and $\boldsymbol{G}(\mathbf{r},\mathbf{r}',\omega)$ the Green tensors of the system in the cases where the matter inside the space region V is present and absent, respectively, with both of them taking into account the bodies in the space region V_{B} in Figs. 3.1 and 3.2. Since the Green tensor $\boldsymbol{G}_V(\mathbf{r},\mathbf{r}',\omega)$ satisfies, for real ω, the relation (A.60) with $\boldsymbol{G}(\mathbf{r},\mathbf{r}',\omega)\mapsto\boldsymbol{G}_V(\mathbf{r},\mathbf{r}',\omega)$, i.e.,

$$\left(\boldsymbol{\nabla}\times\boldsymbol{\nabla}\times-\frac{\omega^2}{c^2}\right)\mathrm{Im}\,\boldsymbol{G}_V(\mathbf{r},\mathbf{r}',\omega) = \mu_0\omega\mathrm{Re}\int \mathrm{d}^3s\,\boldsymbol{Q}(\mathbf{r},\mathbf{s},\omega)\cdot\boldsymbol{G}_V(\mathbf{s},\mathbf{r}',\omega), \tag{3.48}$$

it follows that

$$\boldsymbol{\nabla}\cdot\mathrm{Im}\,\boldsymbol{G}_V(\mathbf{r},\mathbf{r}',\omega) = -\frac{1}{\varepsilon_0\omega}\mathrm{Re}\,\boldsymbol{\nabla}\cdot\int \mathrm{d}^3s\,\boldsymbol{Q}(\mathbf{r},\mathbf{s},\omega)\cdot\boldsymbol{G}_V(\mathbf{s},\mathbf{r}',\omega). \tag{3.49}$$

With the matter inside V taken into account $[\boldsymbol{G}(\mathbf{r},\mathbf{r}',\omega)\mapsto\boldsymbol{G}_V(\mathbf{r},\mathbf{r}',\omega)]$, the expectation value $\langle\underline{\hat{\rho}}(\mathbf{r},\omega)\underline{\hat{\mathbf{E}}}^\dagger(\mathbf{r}',\omega')\rangle$ [Eq. (3.46)] may thus be written as

$$\langle\underline{\hat{\rho}}(\mathbf{r},\omega)\underline{\hat{\mathbf{E}}}^\dagger(\mathbf{r}',\omega')\rangle = -\frac{\hbar\mu_0\omega}{\pi}\,\delta(\omega-\omega')\mathrm{Re}\,\boldsymbol{\nabla}\cdot\int \mathrm{d}^3s\,\boldsymbol{Q}(\mathbf{r},\mathbf{s},\omega)\cdot\boldsymbol{G}_V(\mathbf{s},\mathbf{r}',\omega). \tag{3.50}$$

To calculate in a similar way the quantity $\langle\underline{\hat{\mathbf{j}}}(\mathbf{r},\omega)\times\underline{\hat{\mathbf{B}}}^\dagger(\mathbf{r}',\omega')\rangle$, we note that the (tensorial) Eq. (3.47) implies $[\boldsymbol{G}(\mathbf{r},\mathbf{r}',\omega)\mapsto\boldsymbol{G}_V(\mathbf{r},\mathbf{r}',\omega)]$

$$\begin{aligned}\langle\underline{\hat{\mathbf{j}}}(\mathbf{r},\omega)\times\underline{\hat{\mathbf{B}}}^\dagger(\mathbf{r}',\omega')\rangle &= \frac{\hbar}{\pi}\,\delta(\omega-\omega')\\ &\times\left\{\boldsymbol{\nabla}'\,\mathrm{Tr}\left[\left(\boldsymbol{\nabla}\times\boldsymbol{\nabla}\times-\frac{\omega^2}{c^2}\right)\mathrm{Im}\,\boldsymbol{G}_V(\mathbf{r},\mathbf{r}',\omega)\right]\right.\\ &\qquad\left.-\boldsymbol{\nabla}'\cdot\left(\boldsymbol{\nabla}\times\boldsymbol{\nabla}\times-\frac{\omega^2}{c^2}\right)\mathrm{Im}\,\boldsymbol{G}_V(\mathbf{r},\mathbf{r}',\omega)\right\},\end{aligned} \tag{3.51}$$

which, with Eq. (3.48), becomes

$$\langle\hat{\underline{\mathbf{j}}}(\mathbf{r},\omega)\times\hat{\underline{\mathbf{B}}}^{\dagger}(\mathbf{r}',\omega')\rangle = \frac{\hbar\mu_0\omega}{\pi}\,\delta(\omega-\omega')\\ \times\,\mathrm{Re}\Big[\boldsymbol{\nabla}'\,\mathrm{Tr}\int \mathrm{d}^3s\;\boldsymbol{Q}(\mathbf{r},\mathbf{s},\omega)\cdot\boldsymbol{G}_V(\mathbf{s},\mathbf{r}',\omega)\\ -\boldsymbol{\nabla}'\cdot\int \mathrm{d}^3s\;\boldsymbol{Q}(\mathbf{r},\mathbf{s},\omega)\cdot\boldsymbol{G}_V(\mathbf{s},\mathbf{r}',\omega)\Big]. \tag{3.52}$$

Using Eqs. (3.50) and (3.52), we can now calculate from Eq. (3.43) the dispersion force that acts on a body of volume V_{M} (cf. Figs. 3.1 and 3.2) as

$$\mathbf{F} = \frac{\hbar\mu_0}{\pi}\int_0^\infty \mathrm{d}\omega\,\omega\,\mathrm{Re}\int_{V_{\mathrm{M}}}\mathrm{d}^3r\,\Big[\boldsymbol{\nabla}'\,\mathrm{Tr}\int \mathrm{d}^3s\;\boldsymbol{Q}(\mathbf{r},\mathbf{s},\omega)\cdot\boldsymbol{G}_V(\mathbf{s},\mathbf{r}',\omega)\\ -(\boldsymbol{\nabla}+\boldsymbol{\nabla}')\cdot\int \mathrm{d}^3s\;\boldsymbol{Q}(\mathbf{r},\mathbf{s},\omega)\cdot\boldsymbol{G}_V(\mathbf{s},\mathbf{r}',\omega)\Big]_{\mathbf{r}'\to\mathbf{r}}. \tag{3.53}$$

The second term in Eq. (3.53) is a complete tensor divergence so that for this term the d^3r-integral can be turned into an (in general non-vanishing) surface integral over the boundary ∂V_{M} of the volume V_{M},

$$\mathbf{F} = \frac{\hbar\mu_0}{\pi}\int_0^\infty \mathrm{d}\omega\,\omega\,\mathrm{Re}\Big\{\int_{V_{\mathrm{M}}}\mathrm{d}^3r\,\Big[\boldsymbol{\nabla}'\,\mathrm{Tr}\int \mathrm{d}^3s\;\boldsymbol{Q}(\mathbf{r},\mathbf{s},\omega)\cdot\boldsymbol{G}_V(\mathbf{s},\mathbf{r}',\omega)\Big]_{\mathbf{r}'\to\mathbf{r}}\\ -\int_{\partial V_{\mathrm{M}}}\mathrm{d}\mathbf{a}\cdot\Big[\int \mathrm{d}^3s\;\boldsymbol{Q}(\mathbf{r},\mathbf{s},\omega)\cdot\boldsymbol{G}_V(\mathbf{s},\mathbf{r}',\omega)\Big]_{\mathbf{r}'\to\mathbf{r}}\Big\}. \tag{3.54}$$

Equation (3.54) is an exact and general expression for the (zero temperature) dispersion force, and may be viewed as a far reaching generalization of Eq. (3.5). Clearly, in the case where the matter in V_{M} responds (quasi-)locally, the d^3s-integration in Eq. (3.54) can be restricted to V_{M} as the point $\mathbf{r}$ is located in V_{M}. If furthermore the body is an isolated one (Fig. 3.1), the surface integral contribution in Eq. (3.54) vanishes. Note that both of these statements remain true in an approximate sense also if the medium in V_{M} responds non-locally, if the so-called dielectric approximation is applicable (see Ref. [R10]). In this case, the d^3s-integration in Eq. (3.54) can be restricted to V_{M} and the medium inside V_{M} is treated as having bulk-medium properties, i.e., the conductivity tensor $\boldsymbol{Q}(\mathbf{r},\mathbf{r}',\omega)$ for $\mathbf{r}\in V_{\mathrm{M}}$ is replaced in Eq. (3.54) with the bulk-medium conductivity tensor $\boldsymbol{Q}_{\mathrm{M}}(\mathbf{r}-\mathbf{r}',\omega)$ ($\mathbf{r}\in V_{\mathrm{M}}$) assigned to the medium inside V_{M}. Clearly, the characteristic length of spatial dispersion should be small in comparison with the size of V_{M} for such approximations to be justified.

3.3 Applications

In order to see that Eq. (3.54) really contains Eq. (3.5), let us specialize Eq. (3.54) to the case where the material in V_M is a (not necessarily isolated) locally responding dielectric body with an (isotropic) dielectric susceptibility $\chi_\mathrm{M}(\mathbf{r},\omega)$, leaving unspecified (and thus general) the medium properties outside V_M. For $\mathbf{r}\in V_\mathrm{M}$, we therefore put $\boldsymbol{Q}(\mathbf{r},\mathbf{r}',\omega)$ equal to $-i\omega\varepsilon_0\chi_\mathrm{M}(\mathbf{r},\omega)$ $\boldsymbol{I}\delta(\mathbf{r}-\mathbf{r}')$ in Eq. (3.54) to obtain

$$\mathbf{F} = \frac{\hbar}{2\pi c^2}\int_0^\infty \mathrm{d}\omega\,\omega^2\biggl\{\mathrm{Im}\int_{V_\mathrm{M}} \mathrm{d}^3r\,\chi_\mathrm{M}(\mathbf{r},\omega)\boldsymbol{\nabla}\mathrm{Tr}\,[\boldsymbol{G}_V(\mathbf{r},\mathbf{r}',\omega)]_{\mathbf{r}'\to\mathbf{r}} - 2\,\mathrm{Im}\int_{\partial V_\mathrm{M}} \mathrm{d}\mathbf{a}\cdot\chi_\mathrm{M}(\mathbf{r},\omega)[\boldsymbol{G}_V(\mathbf{r},\mathbf{r}',\omega)]_{\mathbf{r}'\to\mathbf{r}}\biggr\}. \tag{3.55}$$

Here we have used that Tr $\boldsymbol{G}_V(\mathbf{r},\mathbf{r}',\omega)$ is symmetric with respect to $\mathbf{r}$ and $\mathbf{r}'$, due to the reciprocity property of $\boldsymbol{G}_V(\mathbf{r},\mathbf{r}',\omega)$. Further, on recalling the analytic properties of the integrands as functions of (complex) ω, we may employ contour integral techniques to represent Eq. (3.55) in the form of

$$\mathbf{F} = -\frac{\hbar}{2\pi c^2}\int_0^\infty \mathrm{d}\xi\,\xi^2\biggl\{\int_{V_\mathrm{M}} \mathrm{d}^3r\,\chi_\mathrm{M}(\mathbf{r},i\xi)\boldsymbol{\nabla}\mathrm{Tr}\,[\boldsymbol{G}_V(\mathbf{r},\mathbf{r}',i\xi)]_{\mathbf{r}'\to\mathbf{r}} - 2\int_{\partial V_\mathrm{M}} \mathrm{d}\mathbf{a}\cdot\chi_\mathrm{M}(\mathbf{r},i\xi)[\boldsymbol{G}_V(\mathbf{r},\mathbf{r}',i\xi)]_{\mathbf{r}'\to\mathbf{r}}\biggr\}. \tag{3.56}$$

[The transition to imaginary frequencies can be done equally well already in Eq. (3.54)].

As mentioned, in Eqs. (3.53)–(3.56) the coincidence limit $\mathbf{r}'\to\mathbf{r}$ has to be performed in such a way that unphysical self-force contributions are removed. The general Eq. (3.54) suggests that a general technique to define this limit should be based on a family of approximate (regularizing) conductivity tensors $\boldsymbol{Q}^{(\epsilon)}(\mathbf{r},\mathbf{s},\omega)$ that tend to the actual $\boldsymbol{Q}(\mathbf{r},\mathbf{s},\omega)$ as the parameter ϵ goes to zero (say). The approximating $\boldsymbol{Q}^{(\epsilon)}(\mathbf{r},\mathbf{s},\omega)$ should obviously be required to vanish sufficiently strongly at small values of $|\mathbf{r}-\mathbf{s}|$ so as to suppress contributions from a small region around $\mathbf{r}'\simeq\mathbf{r}$ in the d^3s-integrals in Eq. (3.54), where the shape of the suppressed region, whose size goes to zero in the limit, should be in line with any existing symmetry properties of the medium described by $\boldsymbol{Q}(\mathbf{r},\mathbf{r}',\omega)$. For the isotropic locally responding body considered in Eqs. (3.55) and (3.56), a natural procedure is therefore to compute $\boldsymbol{G}_V(\mathbf{r},\mathbf{r}',\omega)_{\mathbf{r}'\to\mathbf{r}}$ by averaging the values of $\boldsymbol{G}_V(\mathbf{r},\mathbf{r}',\omega)$ as $\mathbf{r}'$ varies over a small spherical shell centered at $\mathbf{r}$ and considering the limit where the radius of the shell vanishes. One can show that this procedure effectively amounts to a replacement of the Green tensor $[\boldsymbol{G}_V(\mathbf{r},\mathbf{r}',\omega)]_{\mathbf{r}'\to\mathbf{r}}$ with its scattering part $\boldsymbol{G}_V^{(\mathrm{S})}(\mathbf{r},\mathbf{r},\omega)$ (see, e.g., Ref. [R3]). That is to say, at each point $\mathbf{r}$ in V_M, the Green tensor for the corresponding bulk material must be subtracted from $\boldsymbol{G}_V(\mathbf{r},\mathbf{r}',\omega)$ in the limit $\mathbf{r}'\to\mathbf{r}$. For more general media it may be doubtful if this simple prescription to perform the limit $\mathbf{r}'\to\mathbf{r}$ in Eq. (3.54) remains valid, in particular if the medium in V_M is not an isotropic one. It may be expected, however, that the prescription will remain correct at least for isotropic non-locally responding media treated in the dielectric approximation. (Note that in the dielectric approximation the notion of scattering part of the Green tensor is well-defined just as for locally responding media [R10].)

If the locally responding dielectric body considered in Eqs. (3.55) and (3.56) is an isolated one (cf. Fig. 3.1), the surface integral can be dropped. In this case, we have

$$\mathbf{F} = -\frac{\hbar}{2\pi c^2}\int_0^\infty \mathrm{d}\xi\,\xi^2\int_{V_\mathrm{M}}\mathrm{d}^3r\,\chi_\mathrm{M}(\mathbf{r},i\xi)\boldsymbol{\nabla}\mathrm{Tr}\,[\boldsymbol{G}_V(\mathbf{r},\mathbf{r}',i\xi)]_{\mathbf{r}'\to\mathbf{r}}. \tag{3.57}$$

Clearly, the surface integral must not be dropped in the case where the body is an inner part of a larger dielectric body (cf. Fig. 3.2). Equation (3.57) should be compared with Eq. (3.5), which it generalizes. In contrast to Eq. (3.5), it represents the exact force acting on a locally responding dielectric body of given permittivity, since $\chi_\mathrm{M}(\mathbf{r},i\xi)$ is not restricted to small values anymore. Correspondingly, the Green tensor in Eq. (3.57) is the one that takes the presence of the dielectric body fully into account, whereas in the Green tensor in Eq. (3.5) the presence of the dielectric body is not considered. Hence in contrast to Eq. (3.5), Eq. (3.57) includes in the calculation of the Casimir force that acts on a dielectric body the body's retroaction on the electromagnetic ground-state noise of the residual system.

To make this more explicit, one can expand the full Green tensor $\boldsymbol{G}_V(\mathbf{r},\mathbf{r}',i\xi)$ in powers of $\chi_\mathrm{M}(\mathbf{r},i\xi)$ by using the iterative solution to the Dyson-type integral equation that may be easily derived for it. Inserting the resulting Born series for $\boldsymbol{G}_V(\mathbf{r},\mathbf{r}',i\xi)$ in Eq. (3.56), one obtains the corresponding expansion of the Casimir force $\mathbf{F}$ in powers of $\chi_\mathrm{M}(\mathbf{r},i\xi)$. In particular, truncating this expansion at the term linear in $\chi_\mathrm{M}(\mathbf{r},i\xi)$, i.e., replacing $\boldsymbol{G}_V(\mathbf{r},\mathbf{r}',i\xi)$ with its zeroth-order approximation $\boldsymbol{G}(\mathbf{r},\mathbf{r}',i\xi)$, we simply obtain

$$
\mathbf{F} = -\frac{\hbar}{2\pi c^2}\int_0^\infty \mathrm{d}\xi\,\xi^2 \biggl\{ \int_{V_\mathrm{M}} \mathrm{d}^3r\,\chi_\mathrm{M}(\mathbf{r},i\xi)\boldsymbol{\nabla}\mathrm{Tr}\,[\boldsymbol{G}(\mathbf{r},\mathbf{r}',i\xi)]_{\mathbf{r}'\to\mathbf{r}} - 2\int_{\partial V_\mathrm{M}} \mathrm{d}\mathbf{a}\cdot\chi_\mathrm{M}(\mathbf{r},i\xi)[\boldsymbol{G}(\mathbf{r},\mathbf{r}',i\xi)]_{\mathbf{r}'\to\mathbf{r}} \biggr\}. \tag{3.58}
$$

In this case, the prescription for taking the coincidence limit of the Green tensor simply consists in the replacement $[\boldsymbol{G}(\mathbf{r},\mathbf{r}',i\xi)]_{\mathbf{r}'\to\mathbf{r}} \mapsto \boldsymbol{G}^{(\mathrm{S})}(\mathbf{r},\mathbf{r},i\xi)$, where $\boldsymbol{G}^{(\mathrm{S})}(\mathbf{r},\mathbf{r}',i\xi)$ is the scattering part of the Green tensor $\boldsymbol{G}(\mathbf{r},\mathbf{r}',i\xi)$ in the absence of the dielectric matter in V (cf. Figs. 3.1 and 3.2). Note that if the surface integral can be dropped (i.e., if the case sketched in Fig. 3.1 is considered), then Eq. (3.58) becomes identical with Eq. (3.5). It is worth noting that inclusion in Eq. (3.58) of the higher-order terms of the Born series of $\boldsymbol{G}_V(\mathbf{r},\mathbf{r}',i\xi)$ generates an increasing number of many-body corrections.

Before proceeding, it may be appropriate to comment on the use of different stress tensors in the calculation of dispersion forces. Since Eqs. (3.56) and (3.57) directly follow from the (ground-state) expectation value of the Lorentz force as given by Eq. (3.43), they describe the genuine electromagnetic vacuum force acting on bodies or parts of them, without inclusion of any other forces which may be present in order to compensate for this force. In Ref. [77], the Lorentz-force approach is disputed and it is argued that in the calculation of the force the stress tensor associated with the Lorentz force [Eq. (3.85) together with Eq. (3.86)] should be replaced by Minkowski's stress tensor, because it incorporates additional (mechanical) forces in order to enforce mechanical equilibrium. Unfortunately, it cannot incorporate them in a consistent way, however (see also Ref. [R5]). To see this more explicitly, we first note that if Minkowski's stress tensor were used, then Eq. (3.56) would change to

$$
\mathbf{F}^{(\mathrm{Mink})} = \frac{\hbar}{2\pi c^2}\int_0^\infty \mathrm{d}\xi\,\xi^2 \int_{V_\mathrm{M}} \mathrm{d}^3r\,[\boldsymbol{\nabla}\chi_\mathrm{M}(\mathbf{r},i\xi)]\,\mathrm{Tr}\,[\boldsymbol{G}_V(\mathbf{r},\mathbf{r}',i\xi)]_{\mathbf{r}'\to\mathbf{r}}, \tag{3.59}
$$

which differs from Eq. (3.56) by a surface integral, in general. Hence, the two force formulas agree in the case of an isolated body (cf. Fig. 3.1) where the surface integrals do not contribute to the force and both equations reduce to Eq. (3.57). As we will see in Sec. 3.3.1, application of Eq. (3.57) to an atom in the vicinity of a body gives the correct force that acts on the atom due to the presence of the body and hence, application of Eq. (3.59) also gives the correct force. In the case of a weakly polarizable body, this force results from the sum over the two-atom vdW interactions between the atom under consideration and *all* the atoms of the body, as has been well known for a long time (see, e.g., Ref. [10]). More generally, not only the two-atom interactions but all the many-atom interactions must be taken into account to get the exact force formula, as has been demonstrated on the basis of both microscopic and macroscopic descriptions [19–21] irrespective of whether the body is homogeneous or inhomogeneous. Vice versa, this means that, according to Newton's lex tertia, *each* atom of a homogeneous body must be subject to a force and hence, each group of atoms that represents an *inner* part of the body, must also be subject to a force. In contrast, application of Eq. (3.59) to any inner part of the body leads to the paradoxical result that the force identically vanishes, and the only atoms that are subject to a force are the ones at the surface of the body. Since the use of Minkowski's stress tensor leads to two conflicting results, it is lacking in consistency.

Clearly, similar arguments can also be given when two or more bodies are considered rather than a single atom near a body. It is obvious that the inconsistency problem also appears in classical electrodynamics. In particular, from a careful analysis of the wave propagation in material media as well as a proper interpretation of classical electromagnetic force experiments, it is concluded in Ref. [60] that the (energy-momentum four-tensor associated with the) Lorentz force passes the theoretical and experimental tests and qualifies for a correct description of the energy-momentum properties of the electromagnetic field in macroscopic electrodynamics.

The formulation of the force in terms of the stress tensor as discussed in Sec. 3.2.1 is particularly advantageous in the case of homogeneous (locally responding) material. From Eq. (3.56) it is easily seen that for a homogeneous body that is an inner part of a larger body (cf. Fig. 3.2) one may regard as the stress tensor the expression

$$\boldsymbol{T}(\mathbf{r}) = -\frac{\hbar}{\pi c^2}\int_0^\infty \mathrm{d}\xi\,\xi^2\chi_\mathrm{M}(i\xi)\left\{\tfrac{1}{2}\boldsymbol{I}\,\mathrm{Tr}\,[\boldsymbol{G}_V(\mathbf{r},\mathbf{r}',i\xi)] - \boldsymbol{G}_V(\mathbf{r},\mathbf{r}',i\xi)\right\}_{\mathbf{r}'\to\mathbf{r}}, \tag{3.60}$$

and for an isolated homogeneous body (cf. Fig. 3.1) it follows from Eq. (3.57) that one may employ

$$\boldsymbol{T}(\mathbf{r}) = -\frac{\hbar}{2\pi c^2}\int_0^\infty \mathrm{d}\xi\,\xi^2\chi_\mathrm{M}(i\xi)\boldsymbol{I}\,\mathrm{Tr}\,[\boldsymbol{G}_V(\mathbf{r},\mathbf{r}',i\xi)]_{\mathbf{r}'\to\mathbf{r}}. \tag{3.61}$$

Furthermore, the assumed homogeneity then implies that we may let $[\boldsymbol{G}_V(\mathbf{r},\mathbf{r}',i\xi)]_{\mathbf{r}'\to\mathbf{r}} \mapsto \boldsymbol{G}_V^{(\mathrm{S})}(\mathbf{r},\mathbf{r},i\xi)$ in Eqs. (3.60) and (3.61). Needless to say that replacing the full Green tensor $\boldsymbol{G}_V(\mathbf{r},\mathbf{r}',i\xi)$ as appearing in Eqs.(3.60) and (3.61) with the zeroth-order approximation $\boldsymbol{G}(\mathbf{r},\mathbf{r}',i\xi)$ yields again the Casimir force in the case of weakly polarizable material.

3.3.1 Force on Micro-Objects and Atoms

Since nothing has been said about the spatial extension of the bodies under consideration, the applicability of Eqs. (3.56) and (3.57) ranges from (locally responding) dielectric macro-objects to micro-objects, even including single atoms. In particular, in the case of a dielectric body that may be thought of as consisting of distinguishable (electrically neutral but polarizable) micro-constituents frequently called atoms or molecules within the framework of molecular optics, we may assume the validity of the Clausius–Mossotti relation [47, 78],

$$\begin{aligned}\chi_{\mathrm{M}}(\mathbf{r},\omega) &= \varepsilon_0^{-1}\eta(\mathbf{r})\alpha(\omega)[1-\eta(\mathbf{r})\alpha(\omega)/(3\varepsilon_0)]^{-1}\\ &= \varepsilon_0^{-1}\eta(\mathbf{r})\alpha(\omega)\,[1+\chi_{\mathrm{M}}(\mathbf{r},\omega)/3],\end{aligned} \tag{3.62}$$

where $\alpha(\omega)$ is the polarizability of a single micro-constituent and $\eta(\mathbf{r})$ the number density of the micro-constituents (referred to as atoms in the following). It is worth noting that there is no need here—in contrast to Eq. (3.1)—to regard $\alpha(\omega)$ as being calculated in the lowest (non-vanishing) order of perturbation theory according to Eq. (3.3). One can show that Eq. (3.62) is consistent with the requirement that both $\alpha(\omega)$ and $\chi_{\mathrm{M}}(\mathbf{r},\omega)$ be Fourier transforms of response functions iff

$$\eta(\mathbf{r})\alpha(0)/(3\varepsilon_0) < 1. \tag{3.63}$$

Isolated Micro-Object

Let V_{M} be the small volume of an isolated dielectric micro-object (cf. Fig. 3.1) with a dielectric susceptibility $\chi_{\mathrm{M}}(\omega)$. Assuming that, due to the smallness of V_{M}, the scattering part of the Green tensor can be taken out of the space integral at the (appropriately chosen) position $\mathbf{r}$ of the micro-object, from Eq. (3.57) we derive the force acting on the micro-object to be

$$\mathbf{F} = -V_{\mathrm{M}}\,\frac{\hbar}{2\pi c^2}\int_0^\infty \mathrm{d}\xi\,\xi^2\,\chi_{\mathrm{M}}(i\xi)\boldsymbol{\nabla}\mathrm{Tr}\,\boldsymbol{G}_V^{(\mathrm{S})}(\mathbf{r},\mathbf{r},i\xi), \tag{3.64}$$

which, if the dielectric susceptibility is of the Clausius–Mossotti type so that Eq. (3.62) is valid, reads

$$\mathbf{F} = -V_{\mathrm{M}}\eta\,\frac{\hbar\mu_0}{2\pi}\int_0^\infty \mathrm{d}\xi\,\xi^2\,\alpha(i\xi)\left[1+\tfrac{1}{3}\chi_{\mathrm{M}}(i\xi)\right]\boldsymbol{\nabla}\mathrm{Tr}\,\boldsymbol{G}_V^{(\mathrm{S})}(\mathbf{r},\mathbf{r},i\xi). \tag{3.65}$$

Recall that in the case under study the replacement $[\boldsymbol{G}_V(\mathbf{r},\mathbf{r}',i\xi)]_{\mathbf{r}'\to\mathbf{r}} \mapsto \boldsymbol{G}_V^{(\mathrm{S})}(\mathbf{r},\mathbf{r},i\xi)$ can be made. Equation (3.65), which generalizes Eq. (3.7), differs in two respects from Eq. (3.7). Firstly, its validity is no longer restricted to weakly polarizable matter. Secondly, it takes into account the dependence of the force on the shape of the micro-object.

In contrast to Eq. (3.7), the force as given by Eq. (3.65) includes all-order multi-atom vdW interactions of the micro-object, as may be seen by expanding the Green tensor $\boldsymbol{G}_V(\mathbf{r},\mathbf{r}',i\xi)$ in powers of $\chi_{\mathrm{M}}(i\xi)$ (cf. Ref. [21]). If they are disregarded, Eq. (3.65) reduces to (see Ref. [R6])

$$\mathbf{F} = -V_{\mathrm{M}}\eta\,\frac{\hbar\mu_0}{2\pi}\int_0^\infty \mathrm{d}\xi\,\xi^2\,\alpha(i\xi)\boldsymbol{\nabla}\mathrm{Tr}\,\boldsymbol{G}^{(\mathrm{S})}(\mathbf{r},\mathbf{r},i\xi), \tag{3.66}$$

which, as expected, is nothing but Eq. (3.7)—only the term linear in $\alpha(i\xi)$ contributes to the force. The force in this limit is simply the sum of the forces acting on the atoms due to the presence of the external bodies (region V_{B} in Fig. 3.1). Hence, $\mathbf{F}^{(\mathrm{at})} = (V_{\mathrm{M}}\eta)^{-1}\mathbf{F}$ is the force acting on a single ground-state atom, that is to say, we are left exactly with the formula for the CP force as given by Eq. (3.1), with the exception that now the atomic polarizability is the exact one rather than the perturbative expression given in Eq. (3.4).

Micro-Object That Is an Inner Part of a Larger Body

Let now V_{M} be the small volume of a dielectric micro-object that belongs to a larger body of volume V of the same atoms (cf. Fig. 3.2). Under assumptions analogous to those leading from Eq. (3.57) to Eq. (3.65), from Eq. (3.56) [together with Eq. (3.62)] we obtain the following formula for the (shape-dependent) force acting on the micro-object:

$$\begin{aligned}\mathbf{F} = -V_{\mathrm{M}}\eta\,\frac{\hbar\mu_0}{\pi}\int_0^\infty \mathrm{d}\xi\,\xi^2\,\alpha(i\xi)\left[1+\tfrac{1}{3}\chi_{\mathrm{M}}(i\xi)\right]&\\ \times\ \boldsymbol{\nabla}\cdot\left[\tfrac{1}{2}\boldsymbol{I}\,\mathrm{Tr}\,\boldsymbol{G}_V^{(\mathrm{S})}(\mathbf{r},\mathbf{r},i\xi) - \boldsymbol{G}_V^{(\mathrm{S})}(\mathbf{r},\mathbf{r},i\xi)\right].&\end{aligned} \tag{3.67}$$

Equation (3.67) differs from Eq. (3.65) in the second term in the square brackets in the second line. This difference can be regarded as reflecting the fact that—in contrast to Eq. (3.65)—the force acting on the micro-object is screened by the residual part of the body.

If the multi-atom vdW interactions of the body (of volume V) are disregarded, then Eq. (3.67) can be shown to reduce to the term linear in the atomic polarizability,

$$\mathbf{F} = -V_{\mathrm{M}}\eta\,\frac{\hbar\mu_0}{\pi}\int_0^\infty \mathrm{d}\xi\,\xi^2\,\alpha(i\xi)\boldsymbol{\nabla}\cdot\left[\tfrac{1}{2}\boldsymbol{I}\mathrm{Tr}\,\boldsymbol{G}^{(\mathrm{S})}(\mathbf{r},\mathbf{r},i\xi)-\boldsymbol{G}^{(\mathrm{S})}(\mathbf{r},\mathbf{r},i\xi)\right]. \tag{3.68}$$

Recall that in this approximation $\boldsymbol{G}_V^{(\mathrm{S})}(\mathbf{r},\mathbf{r},i\xi)$ can be replaced with $\boldsymbol{G}^{(\mathrm{S})}(\mathbf{r},\mathbf{r},i\xi)$. From Eq. (3.68) it then follows that $\mathbf{F}^{(\mathrm{at})}=(V_{\mathrm{M}}\eta)^{-1}\mathbf{F}$ can be regarded as the screened CP force acting on an atom of a weakly polarizable medium.

Let us apply Eq. (3.68) to the atoms of a weakly polarizable medium (corresponding to the region V in Fig. 3.2) in front of a laterally infinitely extended magnetodielectric planar wall (corresponding to the region V_{B} in Fig. 3.2), which is assumed to extend from some negative z value up to $z=0$. Using the explicit form of the Green tensor for planar multi-layer structures (see, e.g., Refs. [48,79]), we may write its scattering part for coincident spatial arguments in the (empty) space region $z>0$ as

$$\boldsymbol{G}^{(\mathrm{S})}(\mathbf{r},\mathbf{r},\omega) = \frac{i}{8\pi^2k^2}\int \mathrm{d}^2q\,\frac{e^{2i\beta z}}{\beta}\left\{r_-^p\left[q^2\mathbf{e}_z\mathbf{e}_z-\beta^2\mathbf{e}_q\mathbf{e}_q\right]+r_-^s k^2\mathbf{e}_s\mathbf{e}_s\right\}, \tag{3.69}$$

with $k=k(\omega)=\omega/c$, $q=|\mathbf{q}|$, $\beta=\beta(\omega,q)=(k^2-q^2)^{1/2}$ and orthogonal unit vectors $\mathbf{e}_q=\mathbf{q}/q$, $\mathbf{e}_z=\boldsymbol{\nabla}z$, and $\mathbf{e}_s=\mathbf{e}_q\times\mathbf{e}_z$. The effect of the (multi-layered) wall is described in terms of the generalized reflection coefficients $r_-^\sigma=r_-^\sigma(\omega,q)$ $(\sigma=s,p)$, which in the simplest case of an internally homogeneous, semi-infinite wall reduce to the usual Fresnel amplitudes. From Eq. (3.69) it follows that

$$\mathrm{Tr}\,\boldsymbol{G}^{(\mathrm{S})}(\mathbf{r},\mathbf{r},\omega) = \frac{i}{4\pi}\int_0^\infty \mathrm{d}q\,q\,\frac{e^{2i\beta z}}{\beta}\left[(r_-^s-r_-^p)+\frac{2q^2}{k^2}r_-^p\right]. \tag{3.70}$$

Substitution of Eq. (3.70) into Eq. (3.66) then leads to the well-known expression [54–58] for the CP force acting on a single ground-state atom in front of a planar wall. The screened force acting on an atom of a weakly polarizable medium is obtained by substituting Eqs. (3.69) and (3.70) into Eq. (3.68). The result is ($\beta = i\kappa$)

$$\mathbf{F}^{(\mathrm{at})}(z) = (V_{\mathrm{M}}\eta)^{-1}\mathbf{F}(z) = \mathbf{e}_z \frac{\hbar\mu_0}{4\pi^2}\int_0^\infty \mathrm{d}\xi\, \xi^2\alpha(i\xi)\int_0^\infty \mathrm{d}q\, q e^{-2\kappa z}(r_-^s - r_-^p). \tag{3.71}$$

It fully agrees with the result found by calculating the Casimir stress (3.85) [together with Eq. (3.86)] in a dielectric layer of a planar multi-layer structure and performing therein the limit to weakly polarizable matter (see Refs. [13] and [R4]).

3.3.2 Van der Waals Interaction Between Two Ground-State Atoms

Equation (3.56) can also be regarded as a basic equation for calculating the force between two (ground-state) atoms. For this purpose, let us consider the small change $\delta\mathbf{F}$ of $\mathbf{F}$ in Eq. (3.56) due to a small change $\delta\chi_1(\mathbf{r}, i\xi)$ of the susceptibility $\chi_{\mathrm{M}}(\mathbf{r}, i\xi)$ and a small change $\delta\chi_2(\mathbf{r}, i\xi)$ of the susceptibility $\chi_{\mathrm{B}}(\mathbf{r}, i\xi)$ (of one of the bodies) in the region V_{B} in Fig. (3.2). In particular let us assume that $\chi_{\mathrm{M}}(\mathbf{r}, i\xi)$ only changes inside the region V_{M}. It is not difficult to calculate $\delta\mathbf{F}$ up to second order in $\delta\chi_k(\mathbf{r}, i\xi)$ ($k = 1, 2$) and pick out the term $\delta_{12}\mathbf{F}$ that is bilinear in $\delta\chi_1(\mathbf{r}, i\xi)$ and $\delta\chi_2(\mathbf{r}, i\xi)$:

$$\begin{aligned}\delta_{12}\mathbf{F} = \frac{\hbar}{2\pi c^4}&\int_0^\infty \mathrm{d}\xi\, \xi^4 \int_{V_{\mathrm{M}}} \mathrm{d}^3 r\, \delta\chi_1(\mathbf{r}, i\xi)\\ &\times \int_{V_{\mathrm{B}}} \mathrm{d}^3 s\, \delta\chi_2(\mathbf{s}, i\xi)\boldsymbol{\nabla}\mathrm{Tr}\,[\boldsymbol{G}_V(\mathbf{r}, \mathbf{s}, i\xi)\cdot\boldsymbol{G}_V(\mathbf{s}, \mathbf{r}, i\xi)],\end{aligned} \tag{3.72}$$

where the Green tensor $\boldsymbol{G}_V(\mathbf{r}_1, \mathbf{r}_2, i\xi)$ refers to the system before the susceptibilities have been changed. Note that, since we are dealing with the interaction between two well-separated space regions, the problem of removing 'self'-force contributions does not arise here.

Now let us suppose that the small changes $\delta\chi_1(\mathbf{r}, i\xi)$ and $\delta\chi_2(\mathbf{r}, i\xi)$ result from the introduction into the system of additional atoms, say impurity atoms, of type 1 and type 2, respectively. The (body-assisted) force acting on a type-1 atom at position $\mathbf{r}_1$ due to its interaction with a type-2 atom at position $\mathbf{r}_2$ is then evidently obtained, in first order of their polarizabilities

$\alpha_1(i\xi)$ and $\alpha_2(i\xi)$, from the 'crossing term' $\delta_{12}\mathbf{F}$ as

$$\mathbf{F}_{12}^{(\mathrm{at})} = \frac{\hbar\mu_0^2}{2\pi}\int_0^\infty \mathrm{d}\xi\,\xi^4\alpha_1(i\xi)\alpha_2(i\xi)\boldsymbol{\nabla}_1\mathrm{Tr}\,[\boldsymbol{G}_V(\mathbf{r}_1,\mathbf{r}_2,i\xi)\cdot\boldsymbol{G}_V(\mathbf{r}_2,\mathbf{r}_1,i\xi)], \tag{3.73}$$

which is in full agreement with previous calculations of the vdW interaction between two atoms [59, 80, 81]. Recall that $\boldsymbol{G}_V(\mathbf{r}_1,\mathbf{r}_2,i\xi)$ is the Green tensor for the material system that has been present before the introduction of the additional atoms.

Disregarding local-field corrections, one may insert in Eq. (3.73) the Green tensor for the unperturbed host media. In particular, the force between two atoms embedded in a homogeneous background medium is then obtained by identifying $\boldsymbol{G}_V(\mathbf{r}_1,\mathbf{r}_2,i\xi)$ with the respective bulk-medium Green tensor. Note that in this case the same formula for the force can be obtained by basing the calculations on Minkowski's stress tensor [13, 15, 82]. Choosing in Eq. (3.73) the free-space Green tensor, we recover the vdW interaction between two atoms in otherwise empty space. It should be pointed out that $\mathbf{F}_{12}^{(\mathrm{at})}$ and $\mathbf{F}_{21}^{(\mathrm{at})}$ obey the lex tertia $\mathbf{F}_{12}^{(\mathrm{at})}=-\mathbf{F}_{21}^{(\mathrm{at})}$ if the Green tensor is translationally invariant $[\boldsymbol{G}_V(\mathbf{r}_1+\mathbf{v},\mathbf{r}_2+\mathbf{v},i\xi)=\boldsymbol{G}_V(\mathbf{r}_1,\mathbf{r}_2,i\xi)]$, as it is the case for the two atoms being in bulk material or in free space. Since Eq. (3.73) describes the atom–atom force in the presence of arbitrary macroscopic bodies, it is clear that the atomic positions $\mathbf{r}_1$ and $\mathbf{r}_2$ are not physically equivalent in general.

3.3.3 Casimir Force in Planar Structures

Let us illustrate the theory also for a planar structure composed of locally responding magnetodielectric material, defined according to

$$\varepsilon(\mathbf{r},\omega) = \begin{cases} \varepsilon_-(z,\omega), & z<0,\\ \varepsilon_j(\omega), & 0<z<d_j,\\ \varepsilon_+(z,\omega), & z>d_j, \end{cases} \tag{3.74}$$

$$\mu(\mathbf{r},\omega) = \begin{cases} \mu_-(z,\omega), & z<0,\\ \mu_j(\omega), & 0<z<d_j,\\ \mu_+(z,\omega), & z>d_j. \end{cases} \tag{3.75}$$

It is advantageous to resort to the stress tensor formulation. To determine the Casimir stress in the interspace $0<z<d_j$, we need the corresponding Green

tensor for both spatial arguments within the interspace ($0<z=z'<d_j$). As mentioned, the Green tensor for this geometry is known and can be taken, e.g., from Refs. [48, 79]. Since the transverse projection $\mathbf{q}$ of the wave vector is conserved and the polarizations $\sigma=s,p$ decouple, the scattering part of the Green tensor within the interspace can be expressed in terms of the reflection coefficients $r^\sigma_{j\pm}=r^\sigma_{j\pm}(\omega,q)$ referring to reflection of waves at the right (+) and left (−) wall, respectively, as seen from the interspace. Explicit (recurrence) expressions for the reflection coefficients are available if the walls are multi-slab magnetodielectrics like Bragg mirrors, see, e.g., Refs. [48, 79], [R1]. (For continuous wall profiles, Riccati-type equations have to be solved [48].) As mentioned, in the simplest case of two homogeneous, semi-infinite walls, the coefficients $r^\sigma_{j\pm}$ reduce to the Fresnel amplitudes. In the case first treated by Lifshitz [10], the interspace is empty and the walls are nonmagnetic.

Casimir Stress within a Nonempty Interspace

For the sake of generality, we first leave the wall structure unspecified. By modifying the expression for the scattering part of the Green tensor as given in Ref. [79] to account also for magnetic properties, from Eq. (3.38) together with Eq. (3.39) (without the bulk part of the Green tensor) it then follows that the relevant stress tensor element $T_{zz}(\mathbf{r},\mathbf{r})$ in the interspace $0<z<d_j$ can be given, at zero temperature, in the form of

$$T_{zz}(\mathbf{r},\mathbf{r}) = -\frac{\hbar}{8\pi^2}\int_0^\infty \mathrm{d}\omega\,\mathrm{Re}\int_0^\infty \mathrm{d}q\,q\,\frac{\mu_j(\omega)}{\beta_j(\omega,q)}\,g_j(z,\omega,q) \tag{3.76}$$

($q=|\mathbf{q}|$), where the function $g_j(z,\omega,q)$, which in general depends on the position z within the interspace, reads

$$\begin{aligned} g_j(z,\omega,q) &= 2\big[\beta_j^2(1+n_j^{-2}) - q^2(1-n_j^{-2})\big]D_{js}^{-1} r^s_{j+} r^s_{j-} e^{2i\beta_j d_j} \\ &\quad + 2\big[\beta_j^2(1+n_j^{-2}) + q^2(1-n_j^{-2})\big]D_{jp}^{-1} r^p_{j+} r^p_{j-} e^{2i\beta_j d_j} \\ &\quad - (\beta_j^2+q^2)(1-n_j^{-2})D_{js}^{-1}\big[r^s_{j-}e^{2i\beta_j z} + r^s_{j+}e^{2i\beta_j(d_j-z)}\big] \\ &\quad + (\beta_j^2+q^2)(1-n_j^{-2})D_{jp}^{-1}\big[r^p_{-}e^{2i\beta_j z} + r^p_{j+}e^{2i\beta_j(d_j-z)}\big], \end{aligned} \tag{3.77}$$

with the definitions

$$n_j^2 = n_j^2(\omega) = \varepsilon_j(\omega)\mu_j(\omega), \tag{3.78}$$

$$\beta_j = \beta_j(\omega,q) = (\omega^2 n_j^2/c^2 - q^2)^{1/2}, \tag{3.79}$$

$$D_{j\sigma} = D_{j\sigma}(\omega,q) = 1 - r^\sigma_{j+} r^\sigma_{j-} e^{2i\beta_j d_j}. \tag{3.80}$$

Note that the equations $D_{j\sigma}(\omega,q)=0$ determine, for real $\mathbf{q}$, the frequencies of the guided waves in the planar structure. For practical reasons, it may be advantageous to transform the integral over real frequencies in Eq. (3.76) into an integral along the imaginary frequency axis by means of contour integral techniques [cf. Eqs. (3.38) and (3.40)]. Equation (3.76) is thereby turned into

$$T_{zz}(\mathbf{r},\mathbf{r}) = \frac{\hbar}{8\pi^2}\int_0^\infty \mathrm{d}\xi \int_0^\infty \mathrm{d}q\, q\, \frac{\mu_j(i\xi)}{i\beta_j(i\xi,q)}\, g_j(z,i\xi,q). \tag{3.81}$$

From the derivation it is obvious that the stress formula (3.76) [together with Eq. (3.77)] allows for a locally responding magnetodielectric medium in the interspace. By contrast, Minkowski's stress tensor leads to [23], [R1] ($\mu_j \equiv 1$)

$$T_{zz}^{(\mathrm{M})}(\mathbf{r},\mathbf{r}) = -\frac{\hbar}{2\pi^2}\int_0^\infty \mathrm{d}\omega\, \mathrm{Re}\int_0^\infty \mathrm{d}q\, q\beta_j \sum_{\sigma=s,p} \frac{r_{j+}^\sigma r_{j-}^\sigma e^{2i\beta_j d_j}}{D_{j\sigma}}\,. \tag{3.82}$$

From Eq. (3.77) it is easily seen that for an empty interspace, i.e., $\varepsilon_j=\mu_j=1$, $g_j(z,\omega,q)$ becomes independent of z and simplifies to

$$g_j(z,\omega,q) \to g_j(\omega,q) = 4\beta_j^2 \sum_{\sigma=s,p} \frac{r_{j+}^\sigma r_{j-}^\sigma e^{2i\beta_j d_j}}{D_{j\sigma}}\,. \tag{3.83}$$

In this case, and only in this case, Eq. (3.76) reduces to Eq. (3.82), from which in the case of semi-infinite (homogeneous) dielectric walls Lifshitz's well-known formula [10] can be recovered. As already mentioned, formulas of the type of Eq. (3.82) [which need not necessarily be derived within a stress tensor formulation] have been claimed to apply also to the case where the interspace is filled with dielectric material [11, 24], at least if the material is nonabsorbing [23] (see also the textbooks [5, 15, 22] and references therein). Since $T_{zz}^{(\mathrm{M})}(\mathbf{r},\mathbf{r})$ does not depend on the position z within the interspace, application of Eq. (3.82) implies the very paradoxical result that the force acting on any slice of material selected within the interspace vanishes identically, regardless of the presence and arrangement of the remaining material (in particular, regardless of the yet unspecified walls). This unphysical result clearly shows that Eq. (3.82) cannot be valid if the interspace is not empty, not even if it may be justified to regard the interspace medium as nonabsorbing. In contrast, the stress $T_{zz}(\mathbf{r},\mathbf{r})$ obtained from Eq. (3.76) [together with Eq. (3.77)] is not uniform within an interspace if the interspace is filled with a medium. Hence it gives rise, in general, to a nonvanishing force on a slice of interspace material, and no paradox appears.

Let us return to the stress formula (3.76) [together with Eq. (3.77)]. It is not difficult to see that, for a nonempty interspace, the q-integral in Eq. (3.76) fails to converge at $z=0$ and $z=d_j$, i.e., on the interfaces where the different materials are in immediate contact with each other. Mathematically, the reason for this divergence can be seen in the fact that the reflection coefficients obtained under the assumption of *infinite* lateral extension of the system do not approach zero as q tends to infinity. However, large values of q correspond to waves traveling very obliquely. In any real planar setup of finite lateral extension, such high-q waves clearly do not contribute to the q-integral at all; they are not reflected but walk off instead. Note that a divergence of exactly the same type already appears also in the standard case of an empty interspace in the limit $d_j \to 0$. In order to (approximately) take into account the finite lateral extension of an actual planar setup, an appropriately chosen cutoff value (depending on the lateral system size) for the reflection coefficients at high q values could be introduced, thereby rendering the q-integral finite. Of course, a more satisfactory approach would be to abandon the translational invariance from the outset, which, however, leads to serious mathematical difficulties since waves with different polarizations and transverse wave vectors are then no longer decoupled.

Since the Casimir force acting on a body is given by the integral of the stress tensor over the surface enclosing the body, the stress tensor on its own is of less importance. What is really important is the integral force value. To obtain the force (per unit area) acting on a (multilayered) plate of infinite lateral extension, the stress on the two sides of the plate must be taken into account. As the example given below shows, it may then happen that the parts of the stress tensor that diverge when the plate is approached from the two sides cancel each other out. In such a case, the Casimir force (per unit area) on a plate remains well defined even if its lateral extension is assumed to be infinite.

Casimir Force on a Plate in a Nonempty Cavity

In order to make contact with recent work on the Casimir force on bodies embedded in media [23], let us calculate the force acting at zero temperature on a homogeneous plate in a nonempty planar cavity, according to the five-region setup as sketched in Fig. 3.3. The cavity walls are labeled by $l=0$

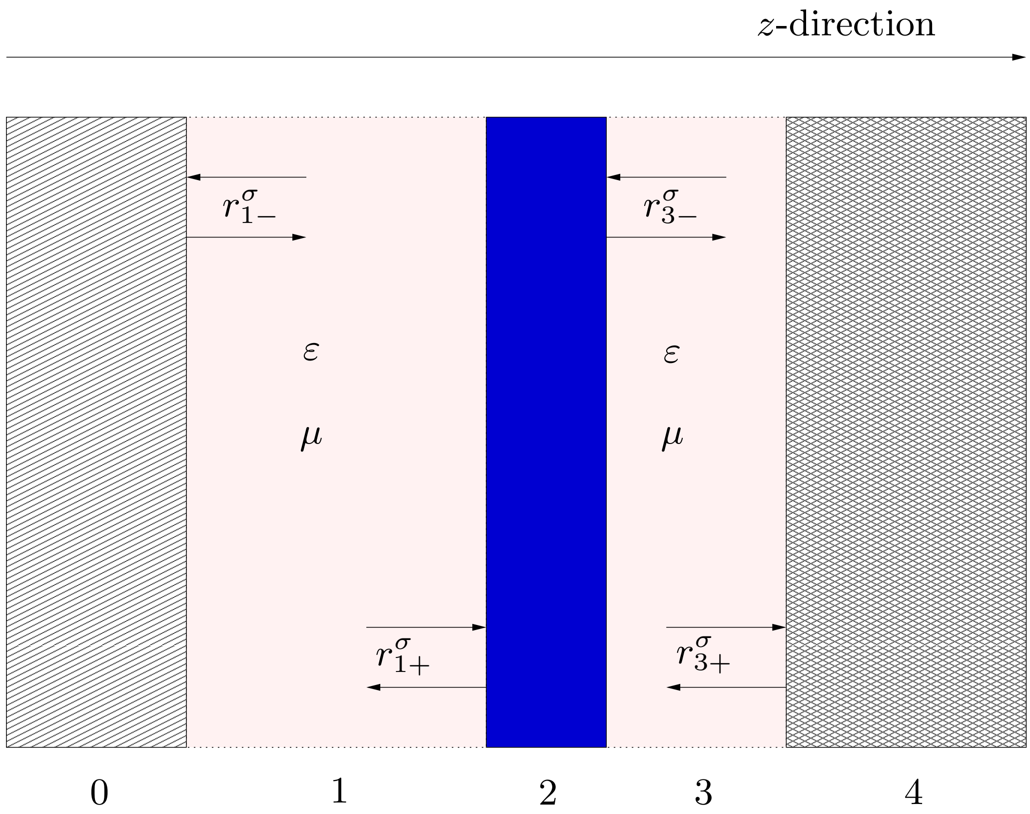

Figure 3.3: Homogeneous plate embedded in a nonempty cavity. The cavity medium on the right and left sides of the plate is the same.

and $l=4$, the plate by $l=2$, and the cavity regions that are filled with the medium the plate is embedded in are labeled by $l=1$ and $l=3$, with $\varepsilon(\omega)\equiv\varepsilon_1(\omega)=\varepsilon_3(\omega)$ and $\mu(\omega)\equiv\mu_1(\omega)=\mu_3(\omega)$. The total (volume) force per unit transverse area acting on the plate can be obtained by (vectorial) addition of the two force contributions from the two sides of the plate. Application of Eq. (3.81) then yields the total force per unit transverse area in the form of

$$F=\frac{\hbar}{8\pi^2}\int_0^\infty\mathrm{d}\xi\int_0^\infty\mathrm{d}q\,q\,\frac{\mu(i\xi)}{i\beta(i\xi,q)}\left[g_3(0,i\xi,q)-g_1(d_1,i\xi,q)\right] \tag{3.84}$$

$[\beta(i\xi,q)\equiv\beta_1(i\xi,q)=\beta_3(i\xi,q)]$.

For a quantitative comparison with specific results obtained in Ref. [23] on the basis of Minkowski's stress tensor, we make the following simplifying assumptions. We assume that (i) all the reflection coefficients can be regarded as being almost constant, and (ii) the reflection coefficients r_{1+}^{σ} and

r^σ_{3-} can be approximated by the (same) single-interface (Fresnel) reflection coefficient $r^\sigma_{1/2}$. Physically, these assumptions mean that (i) the distances d_1 and d_3 between the plate and the cavity walls must not be too small, and (ii) the plate must be thick enough. Moreover, the approximation scheme implies that the permittivity and the permeability of the medium the plate is embedded in can be replaced with their static values briefly referred to as ε and μ in the following, with $n=\sqrt{\varepsilon\mu}$ being the static refractive index. From Eq. (3.77) it then follows that the difference of the functions $g_3(0,i\xi,q)$ and $g_1(d_1,i\xi,q)$ appearing in Eq. (3.84) can be approximated according to

$$\begin{aligned} g_3(0,i\xi,q)-g_1(d_1,i\xi,q) \quad &\simeq \sum_{\sigma=s,p}\Bigg\{2\left(\frac{1}{D_{3\sigma}}-\frac{1}{D_{1\sigma}}\right)\left[\beta^2\left(1+\frac{1}{n^2}\right)\right. \\ &\left.+\Delta_\sigma q^2\left(1-\frac{1}{n^2}\right)\right]+\Delta_\sigma(\beta^2+q^2)\left(1-\frac{1}{n^2}\right) \\ &\times\left[\frac{r^\sigma_{1/2}+r^\sigma_{3+}e^{2i\beta d_3}}{D_{3\sigma}}-\frac{r^\sigma_{1/2}+r^\sigma_{1-}e^{2i\beta d_1}}{D_{1\sigma}}\right]\Bigg\} \end{aligned} \tag{3.85}$$

$(\Delta_\sigma=\delta_{\sigma p}-\delta_{\sigma s})$, where

$$\frac{r^\sigma_{3+}e^{2i\beta d_3}}{D_{3\sigma}}-\frac{r^\sigma_{1-}e^{2i\beta d_1}}{D_{1\sigma}}=\frac{1-D_{3\sigma}}{r^\sigma_{3-}D_{3\sigma}}-\frac{1-D_{1\sigma}}{r^\sigma_{1+}D_{1\sigma}}\simeq\frac{1}{r^\sigma_{1/2}}\left(\frac{1}{D_{3\sigma}}-\frac{1}{D_{1\sigma}}\right). \tag{3.86}$$

[One can carry out the derivations also with the exact (yet more complicated) version of Eq. (3.85), see Ref. [R3].] Substituting Eq. (3.85) together with Eq. (3.86) into Eq. (3.84), we (approximately) obtain

$$\begin{aligned} F=&\frac{\hbar}{8\pi^2}\int_0^\infty\mathrm{d}\xi\int_0^\infty\mathrm{d}q\,q\,\frac{\mu}{i\beta}\sum_{\sigma=s,p}\left(\frac{1}{D_{3\sigma}}-\frac{1}{D_{1\sigma}}\right) \\ &\times\left\{2\beta^2\left(1+\frac{1}{n^2}\right)-\Delta_\sigma\frac{\xi^2}{c^2}\left(n^2-1\right)\left(r^\sigma_{1/2}+\frac{1}{r^\sigma_{1/2}}\right)+2\Delta_\sigma q^2\left(1-\frac{1}{n^2}\right)\right\}. \end{aligned} \tag{3.87}$$

From an inspection of Eq. (3.87) it is seen that there is no divergence; the integrals are well-defined. It is worth noting that even without application of the approximation scheme, the integrals in the basic formula (3.84) do not diverge. The reason is that, for a chosen value of ξ, the coefficients

$r^\sigma_{3-}(i\xi,q)$ and $r^\sigma_{1+}(i\xi,q)$ tend exponentially to the same single-interface Fresnel coefficient $r^\sigma_{1/2}(i\xi,q)$ as q goes to infinity, as may be seen from relations like

$$r^\sigma_{1+} = \frac{r^\sigma_{1/2} + e^{2i\beta_2 d_2} r^\sigma_{2+}}{1 + r^\sigma_{1/2} e^{2i\beta_2 d_2} r^\sigma_{2+}} \to r^\sigma_{1/2} \text{ if } q \to \infty, \tag{3.88}$$

$$r^\sigma_{3-} = \frac{r^\sigma_{3/2} + e^{2i\beta_2 d_2} r^\sigma_{2-}}{1 + r^\sigma_{3/2} e^{2i\beta_2 d_2} r^\sigma_{2-}} \to r^\sigma_{3/2} \text{ if } q \to \infty \tag{3.89}$$

together with the relation $r^\sigma_{3/2} = r^\sigma_{1/2}$ (valid for arbitrary values of ξ and q). Note that $i\beta_2 \to -\infty$ if $q \to \infty$. As a consequence, the divergent contributions to the q-integral in Eq. (3.84), which would arise from $g_3(0,i\xi,q)$ and $g_1(d_1,i\xi,q)$ separately, combine in a convergent fashion. Thus, for the setup under study, a q-cutoff need not be introduced.

Let us return to Eq. (3.87). If the two walls and the plate are almost perfectly reflecting, i.e., $r^\sigma_{1-} \simeq r^\sigma_{3+} \simeq \Delta_\sigma$, $r^\sigma_{1/2} \simeq \Delta_\sigma$, then standard evaluation of the integrals leads to ($n = \sqrt{\varepsilon\mu}$)

$$F = \frac{\hbar c \pi^2}{240} \sqrt{\frac{\mu}{\varepsilon}} \left(\frac{2}{3} + \frac{1}{3\varepsilon\mu}\right) \left(\frac{1}{d_3^4} - \frac{1}{d_1^4}\right). \tag{3.90}$$

In particular, if only one wall is present, say the left one, then Eq. (3.90) reduces to ($d_3 \to \infty$, $d_1 \equiv d$)

$$F = -\frac{\hbar c \pi^2}{240} \sqrt{\frac{\mu}{\varepsilon}} \left(\frac{2}{3} + \frac{1}{3\varepsilon\mu}\right) \frac{1}{d^4}, \tag{3.91}$$

which is the generalization of Casimir's well known formula [83] for the force between two almost perfectly reflecting plates separated by vacuum [$\mu = \varepsilon = 1$ in Eq. (3.91)] to the case where the interspace between the plates is filled with a medium having static permeability μ and static permittivity ε.

In order to compare Eq. (3.90) with the force formula obtained on the basis of Minkowski's stress tensor, we note that the use of Minkowski's stress tensor for a nonmagnetic medium leads to [see Eqs. (3.6) and (3.7) in Ref. [23]]

$$F^{(\mathrm{M})} = -\frac{\hbar}{\pi^2} \int_0^\infty \mathrm{d}\xi \int_0^\infty \mathrm{d}q\, q\, i\beta \left(\frac{1}{e^{-2i\beta d_3} - 1} - \frac{1}{e^{-2i\beta d_1} - 1}\right) \tag{3.92}$$

in place of Eq. (3.87) with $\mu = 1$. For an almost perfectly reflecting plate in a cavity with almost perfectly reflecting walls, standard evaluation of the integrals in Eq. (3.92) then yields, in place of Eq. (3.90),

$$F^{(\mathrm{M})} = \frac{\hbar c \pi^2}{240} \frac{1}{\sqrt{\varepsilon}} \left(\frac{1}{d_3^4} - \frac{1}{d_1^4}\right), \tag{3.93}$$

which in the limit $d_3 \to \infty$ reduces to $(d_1 \equiv d)$

$$F^{(\mathrm{M})} = -\frac{\hbar c \pi^2}{240} \frac{1}{\sqrt{\varepsilon}} \frac{1}{d^4} \,. \tag{3.94}$$

Note that Eq. (3.94) corresponds to the result derived in Ref. [84] by means of mode summation methods [i.e., by resorting to Eq. (1.1)]. Comparing Eq. (3.90) with Eq. (3.93) [or Eq. (3.91) with Eq. (3.94)], we see that

$$|F| \leq |F^{(\mathrm{M})}|, \tag{3.95}$$

i.e., the absolute value of the force is $(n > 1)$ always smaller than that predicted from Minkowski's stress tensor. Introduction of a (polarizable) medium into the interspace is obviously associated with some screening of the plate, thereby reducing the force acting on it. Since the charges and currents attributed to the interspace medium are not correctly treated in a theory that is based on Minkowski's stress tensor or an equivalent formalism, the screening effect is underestimated and consequently the force calculated in this way is overestimated. Although the assumptions made to derive the results given above are rather restrictive, the comparison of Eq. (3.90) with Eq. (3.93) clearly shows that the correct inclusion of the medium into the theory can give rise to noticeable effects (see Fig. 3.4).

A consequence of the approximation scheme employed in this section is the appearance of the (real) values of the static permittivity and the static permeability of the interspace material in Eq. (3.90). However, the basic equation (3.84) is of course valid for arbitrary locally responding magnetodielectric media. The influence of material dispersion and absorption comes into play when the distances d_1 and/or d_3 are decreased. The behavior of the permeability and the permittivity at nonzero frequencies becomes then important.

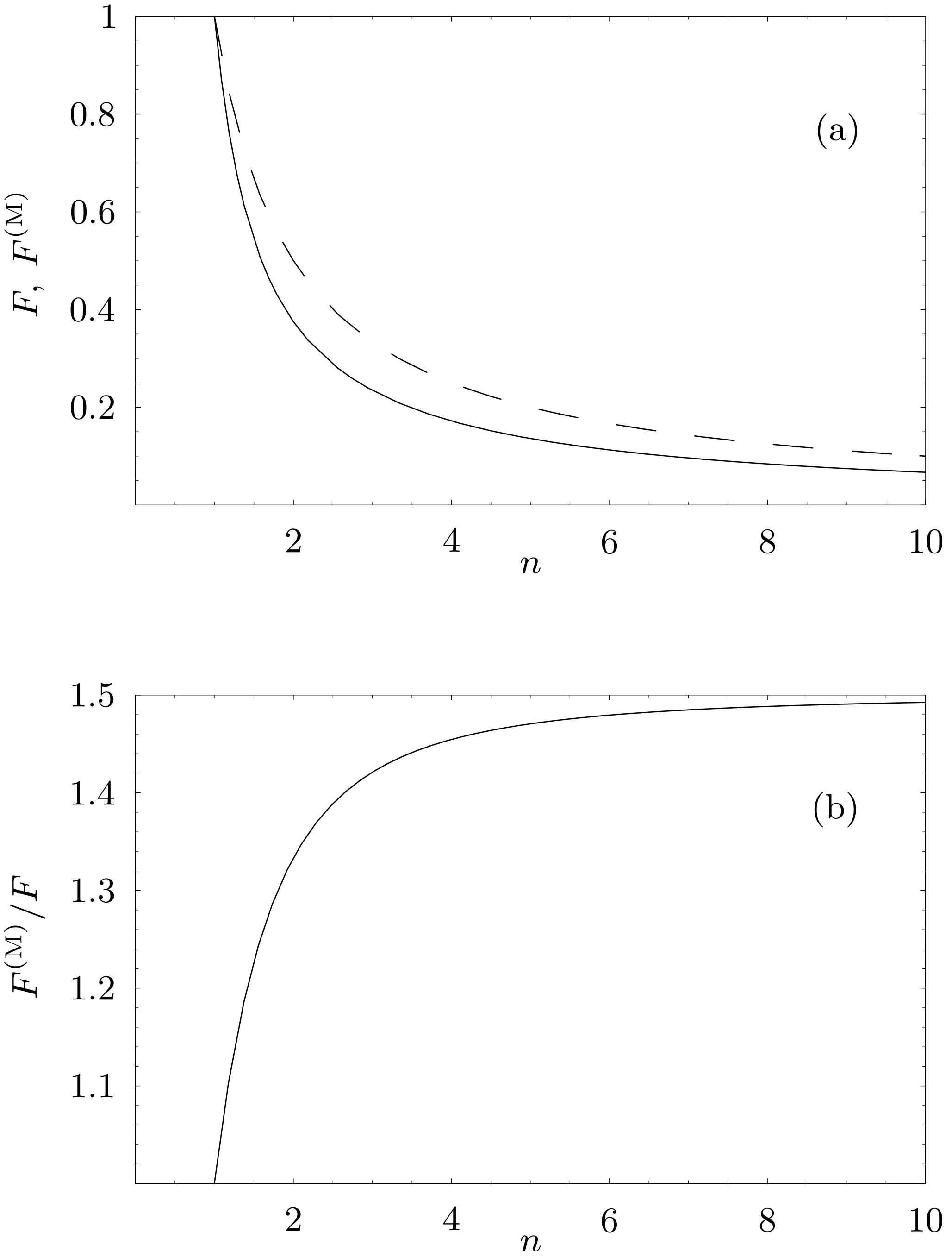

Figure 3.4: (a) The Casimir force F given by Eq. (3.90) (solid curve) is shown as a function of the medium refractive index $n=\sqrt{\varepsilon}$ ($\mu=1$) for chosen distances d_1 and d_3. For comparison, $F^{(\mathrm{M})}$ given by Eq. (3.93) (dashed curve) is shown. (b) The ratio $F^{(\mathrm{M})}/F$ is shown as a function of the medium refractive index.

Chapter 4

Summary

In this work, we have presented a general quantization scheme for the macroscopic electromagnetic field in arbitrary linearly responding media (at rest), thereby offering a unified approach to QED in such media. Describing the medium response by a non-local conductivity tensor, any of the possible electromagnetic features of a linear medium is covered by the scheme, in particular, spatial dispersion. Central quantities of the scheme are the noise current density that is intimately connected with the absorption necessarily observed in any linear medium in equilibrium, the bosonic dynamical variables associated with the noise current density, and the Green tensor of the macroscopic Maxwell equations, in which the medium properties enter via the conductivity tensor. Inclusion in the theory of additional atomic sources that interact with the medium-assisted electromagnetic field may be straightforwardly performed along standard lines, where in this context the 'free' field already incorporates the interaction with some background material. With some modifications, the theory is also capable of handling (linearly) amplifying media in a consistent manner, although the concept of linear amplification strictly speaking does not really fit into the usual linear-response framework.

From a careful analysis of the dynamical variables and (quasi-)local limiting forms of the non-local conductivity tensor, we have shown how quantization schemes previously developed for locally responding media can be recovered as special applications of the general quantization scheme. In particular, a locally responding magnetodielectric medium can be viewed as a special quasi-local limiting case of an isotropic, spatially dispersive medium without optical activity, where the (local) dielectric permittivity and magnetic permeability are just two contributions to one and the same quasi-local conductivity tensor. As a result, application of the general quantization scheme shows that the electromagnetic field in such a medium can be quantized by using a single set of bosonic variables. Generally, the use of a single set of bosonic variables means that the noise current density that enters the macroscopic Maxwell equations is not divided into parts (associated, e.g., with a polarization and a magnetization) regarded as representing independent degrees of freedom, but is rather treated as an entity. This may be particularly advantageous for future studies of (quantum) electrodynamics in moving media, simplifying the discussion of transformations to different frames of reference. However, the theory also admits, by appropriate projection, the use of several independent sets of bosonic variables, which in fact corresponds to the neglect of certain kinds of interactions in the sense of super-selection rules.

Applying the general quantization scheme, we have then presented a unified, macroscopic theory of dispersion forces, which we regard as the purely electromagnetic, fluctuation-induced Lorentz forces that act on the linearly responding current (and corresponding charges) that define a material body within the framework of macroscopic (linear) electrodynamics. We have shown that this viewpoint unites Casimir, Casimir–Polder and van der Waals forces in a very natural way that makes transparent their common physical basis. Dispersion forces acting on (ground-state) macro- and micro-objects—including single atoms—may thus be calculated in a unified way. Other force contributions that in practice may compensate the genuine dispersion forces are not included in the calculation. The Lorentz-force approach may on the other hand be viewed as just providing the necessary size of such additional forces if force compensation is to be assumed. In contrast, approaches that resort to Minkowski's (rather than Maxwell's) stress tensor or related quantities have the aim to include such forces, and in some way demand force compensation from the very beginning. Unfortunately, inconsistencies are thereby introduced. One may expect that a consistent treatment of such additional forces together with the genuine dispersion forces is not possible without detailed microscopic model assumptions about the bodies involved.

On the basis of the Lorentz force, we have derived general formulas for the dispersion force acting on bodies or a parts of them—formulas that apply to arbitrary (linearly responding) media and whose validity is not restricted to weakly polarizable matter. In particular, if the matter may be regarded as consisting of atoms in the broadest sense of the word and a (local) permittivity of Clausius–Mossotti-type can be assigned to it, then all the relevant many-atom vdW interactions of the involved matter can be included in the force to be calculated. As already mentioned, the applicability of the theory ranges from macro-objects to micro-objects. The force acting on a (locally responding) dielectric micro-object is often calculated in the spirit of a simple superposition of CP forces acting on independent atoms, at least in the case of a weakly polarizable object. The present theory enables one to systematically include in the calculation both the dependence of the force on the shape of the micro-object and, at the same time, the contributions to the force due to many-atom interactions of atoms of the micro-object, without restriction to weakly polarizable matter. If the micro-object reduces to a single atom, the well-known formula for the CP force on a single atom is recovered. It is worth noting that not only the force acting on an isolated atom can be obtained, but also the force on a medium atom. For a medium atom, the CP force is screened due to the presence of neighboring medium atoms, while there is of course no such screening in the case of an isolated atom. Moreover, the basic formulas can also be used to study the body-assisted vdW interaction between atoms.

Specializing to planar structures, we have generalized Lifshitz-type formulas for the Casimir force on planar plates (being valid for empty interspaces between the plates) to the case where the interspaces are filled with a (locally responding) magnetodielectric medium. The problems implied by basing the calculation of the Casimir force on Minkowski's stress tensor can in this case be exhibited rather explicitly. (Interestingly, Lifshitz himself did not address nonempty interspaces in his seminal article [10].) Studying the Casimir force acting on a homogeneous plate embedded in a medium in a planar cavity and applying approximations such as high reflection, we have also given the correct extension of Casimir's original formula for the force between two perfectly reflecting plates to the case where the interspace between the plates is filled with a (locally responding magnetodielectric) medium. It shows very clearly that if the plate is embedded in a medium, then the force can noticeably differ from the result obtained on the basis of Minkowski's stress tensor.

Summarizing, we have in this work presented and worked out in detail (i) a universally applicable and versatile theoretical framework for macroscopic QED in arbitrary linearly responding media and (ii) a unified approach to the theory of dispersion forces on ground-state objects (or thermally excited ones). The dispersion force acting on a macroscopic piece of matter may basically be viewed as being 'just' the (quantum) Lorentz force on the constituting charges and currents, which, within the macroscopic description, are completely specified through the linear-response constitutive relation that has been adopted to describe the matter. We think this is a conceptually straightforward and quite satisfactory point of view.

Appendix A

Supplementary Material

A.1 Electro- and Magnetostatics as Limiting Cases

The solution to (the classical) Eq. (2.7) is

$$\underline{\mathbf{E}}(\mathbf{r},\omega) = i\mu_0\omega \int \mathrm{d}^3r' \, \boldsymbol{G}(\mathbf{r},\mathbf{r}',\omega)\cdot\underline{\mathbf{j}}(\mathbf{r}',\omega), \tag{A.1}$$

where $\boldsymbol{G}(\mathbf{r},\mathbf{r}',\omega)$ is the free-space Green tensor, which obeys Eq. (2.18) with $\boldsymbol{Q}(\mathbf{r},\mathbf{r}',\omega)$ replaced with zero (in a limiting sense; compare the remarks at the end of Sec. 2.1). It has the property of not mixing the spaces of longitudinal and transverse vector functions, as may be seen from its straightforward Fourier representation [cf. Eq. (2.37)]

$$\boldsymbol{G}(\mathbf{r},\mathbf{r}',\omega) = \boldsymbol{G}^{\parallel}(\mathbf{r},\mathbf{r}',\omega) + \boldsymbol{G}^{\perp}(\mathbf{r},\mathbf{r}',\omega), \tag{A.2}$$

$$\boldsymbol{G}^{\parallel}(\mathbf{r},\mathbf{r}',\omega) = -\int \frac{\mathrm{d}^3k}{(2\pi)^3}\, e^{i\mathbf{k}\cdot(\mathbf{r}-\mathbf{r}')}\frac{\mathbf{kk}}{k^2}\,\frac{1}{\omega^2/c^2} = -\frac{c^2}{\omega^2}\boldsymbol{\Delta}_{\parallel}(\mathbf{r}-\mathbf{r}'), \tag{A.3}$$

$$\boldsymbol{G}^{\perp}(\mathbf{r},\mathbf{r}',\omega) = \int \frac{\mathrm{d}^3k}{(2\pi)^3}\, e^{i\mathbf{k}\cdot(\mathbf{r}-\mathbf{r}')}\left(\boldsymbol{I}-\frac{\mathbf{kk}}{k^2}\right)\frac{1}{k^2-\omega^2/c^2}. \tag{A.4}$$

Let us consider some slowly varying, nearly static charge density in free space. In the strictly static limit, its frequency components behave like $\underline{\rho}(\mathbf{r},\omega) \to \rho_0(\mathbf{r})\delta(\omega)$. To $\underline{\rho}(\mathbf{r},\omega)$ we may assign a polarization, $\underline{\rho}(\mathbf{r},\omega) = -\boldsymbol{\nabla}\cdot\underline{\mathbf{P}}(\mathbf{r},\omega)$, where correspondingly $\underline{\mathbf{P}}(\mathbf{r},\omega)\to\mathbf{P}_0(\mathbf{r})\delta(\omega)$ in the static limit. From Eq. (2.6), the total current density $\underline{\mathbf{j}}(\mathbf{r},\omega)$ therefore contains the polarization current density $-i\omega\underline{\mathbf{P}}(\mathbf{r},\omega)$, which however vanishes in the static limit $[\omega\delta(\omega)=0]$. In order to calculate the electric field associated with this polarization current in the static limit, we insert $\underline{\mathbf{j}}(\mathbf{r},\omega)\mapsto -i\omega\underline{\mathbf{P}}(\mathbf{r},\omega)$ in Eq. (A.1) and afterwards let $\underline{\mathbf{P}}(\mathbf{r},\omega)\to\mathbf{P}_0(\mathbf{r})\delta(\omega)$ in the limit. Introducing the Coulomb Green function $g(r)=(4\pi r)^{-1}$, taking Eqs. (A.2)–(A.4) into account, and converting to the time-domain, the result is

$$\begin{aligned}\mathbf{E}_{\mathrm{es}}(\mathbf{r},t) &= \int \mathrm{d}\omega\, e^{-i\omega t}\underline{\mathbf{E}}(\mathbf{r},\omega) = \mu_0 \int \mathrm{d}^3r'\, \left[\omega^2 e^{-i\omega t}\boldsymbol{G}(\mathbf{r},\mathbf{r}',\omega)\right]_{\omega\to 0}\cdot\mathbf{P}_0(\mathbf{r}') \\ &= -\varepsilon_0^{-1}\boldsymbol{\nabla}\int \mathrm{d}^3r'\, g(|\mathbf{r}-\mathbf{r}'|)\rho_0(\mathbf{r}') \end{aligned}\tag{A.5}$$

$[\boldsymbol{\Delta}_{\|}(\mathbf{r})=-\boldsymbol{\nabla}\boldsymbol{\nabla}g(r)]$. From an analogous calculation, the contribution of the polarization current to the magnetic induction field is found to be zero in the static limit [cf. Eq. 2.2],

$$\begin{aligned}\mathbf{B}_{\mathrm{es}}(\mathbf{r},t) &= \int \mathrm{d}\omega\, e^{-i\omega t}\underline{\mathbf{B}}(\mathbf{r},\omega) \\ &= -i\mu_0 \int \mathrm{d}^3r'\, \left[\omega\, e^{-i\omega t}\boldsymbol{\nabla}\times\boldsymbol{G}(\mathbf{r},\mathbf{r}',\omega)\right]_{\omega\to 0}\cdot\mathbf{P}_0(\mathbf{r}') = 0. \end{aligned}\tag{A.6}$$

Equation (A.5) is the well-known solution to the equations of electrostatics, $\boldsymbol{\nabla}\cdot\mathbf{E}(\mathbf{r})=\rho_0(\mathbf{r})/\varepsilon_0$, $\boldsymbol{\nabla}\times\mathbf{E}(\mathbf{r})=0$, and Eq. (A.6) states that no static magnetic field is generated from a static polarization.

The total current density may have a further contribution that does not vanish in the static limit but whose frequency components approach $\mathbf{j}_0(\mathbf{r})\delta(\omega)$ with some non-zero $\mathbf{j}_0(\mathbf{r})$, which must be transverse for consistency [cf. Eq. (2.6)], $\mathbf{j}_0(\mathbf{r})=\mathbf{j}_0^{\perp}(\mathbf{r})$. The fields attributed to such a transverse static current may be derived analogously, where $\boldsymbol{G}^{\|}(\mathbf{r},\mathbf{r}',\omega)$ is not needed in the calculation. Noting that $\boldsymbol{G}^{\perp}(\mathbf{r},\mathbf{r}',\omega)$, remains well-defined at zero frequency [see Eq. (A.4)], one finds

$$\mathbf{E}_{\mathrm{ms}}(\mathbf{r},t) = \int \mathrm{d}\omega\, e^{-i\omega t}\underline{\mathbf{E}}(\mathbf{r},\omega) = i\mu_0 \int \mathrm{d}^3r'\, \left[\omega e^{-i\omega t}\boldsymbol{G}^{\perp}(\mathbf{r},\mathbf{r}',\omega)\right]_{\omega\to 0}\cdot\mathbf{j}_0(\mathbf{r}') = 0 \tag{A.7}$$

and

$$\begin{aligned}\mathbf{B}_{\mathrm{ms}}(\mathbf{r},t) &= \int \mathrm{d}\omega\, e^{-i\omega t}\underline{\mathbf{B}}(\mathbf{r},\omega) = \mu_0\boldsymbol{\nabla}\times \int \mathrm{d}^3r'\, \left[e^{-i\omega t}\boldsymbol{G}(\mathbf{r},\mathbf{r}',\omega)\right]_{\omega\to 0}\cdot\mathbf{j}_0(\mathbf{r}') \\ &= \mu_0\boldsymbol{\nabla}\times\int \mathrm{d}^3r'\, g(|\mathbf{r}-\mathbf{r}'|)\boldsymbol{I}\cdot\mathbf{j}_0(\mathbf{r}'). \end{aligned}\tag{A.8}$$

Equation (A.8) is the well-known solution to the equations of magnetostatics, $\boldsymbol{\nabla}\cdot\mathbf{B}(\mathbf{r})=0$, $\boldsymbol{\nabla}\times\mathbf{B}(\mathbf{r})=\mu_0\mathbf{j}_0(\mathbf{r})$, and Eq. (A.7) states that no static electric field is generated by a static (transverse) current. Both electrostatics and magnetostatics have thus been fully recovered from the solution to Eq. (2.7). [Note that the condition $\mathbf{j}_0(\mathbf{r})=\mathbf{j}_0^{\perp}(\mathbf{r})$ is just the integrability condition without which magnetostatics were not consistent.] Since $\rho_0(\mathbf{r})$ and $\mathbf{j}_0(\mathbf{r})$ can be specified independently, electrostatic [Eqs. (A.5), (A.6)] and magnetostatic [Eqs. (A.7), (A.8)] fields may coexist without any interrelation, as is well-known.

A.2 Consistency at Zero Frequency

The analysis of App. A.1, though based on properties of the free-space Green tensor, may be used to obtain physical consistency requirements for the low-frequency behavior of general Green tensors that obey Eq. (2.18) with some general conductivity tensor $\boldsymbol{Q}(\mathbf{r},\mathbf{r}',\omega)$. In contrast to App. A.1, it is now necessary to distinguish between longitudinal and transverse parts of a tensor from the left and from the right. Obviously, it is possible to regard the quantities $\rho_0(\mathbf{r})$ and/or $\mathbf{j}_0(\mathbf{r})$ considered in App. A.1 as arising, partly or completely, from the linear response of a medium in the static limit, as it is encoded in the low-frequency behavior of the corresponding conductivity tensor. In order that [the polarization $\mathbf{P}_0(\mathbf{r})$ attributed to] the static charge density $\rho_0(\mathbf{r})$ and the static current density $\mathbf{j}_0(\mathbf{r})$ remain well-defined also if they are viewed as containing such medium-response contributions, the responsible conductivity tensor has to approach a (zero or non-zero) static limit according to [cf. Eq. (2.29)]

$$\boldsymbol{Q}(\mathbf{r},\mathbf{r}',\omega) \simeq \boldsymbol{Q}_0(\mathbf{r},\mathbf{r}') - i\omega\varepsilon_0\boldsymbol{\chi}_0(\mathbf{r},\mathbf{r}') + \cdots, \tag{A.9}$$

with certain real tensors $\boldsymbol{Q}_0(\mathbf{r},\mathbf{r}')$ and $\boldsymbol{\chi}_0(\mathbf{r},\mathbf{r}')$. The first and second terms of Eq. (A.9) affect magnetostatics and electrostatics, respectively. Since a non-zero static current density must be transverse (cf. App. A.1), it follows that the static conductivity tensor $\boldsymbol{Q}_0(\mathbf{r},\mathbf{r}')$ must generally be transverse from the right, and—due to the reciprocity of $\boldsymbol{Q}(\mathbf{r},\mathbf{r}',\omega)$—it has to be also transverse from the left, i.e.,

$$\int \mathrm{d}^3s\, \boldsymbol{Q}_0(\mathbf{r},\mathbf{s})\cdot\boldsymbol{\Delta}_\perp(\mathbf{s}-\mathbf{r}') = \boldsymbol{Q}_0(\mathbf{r},\mathbf{r}') = \int \mathrm{d}^3s\, \boldsymbol{\Delta}_\perp(\mathbf{r}-\mathbf{s})\cdot\boldsymbol{Q}_0(\mathbf{s},\mathbf{r}'). \tag{A.10}$$

The static fields attributed to $\rho_0(\mathbf{r})$ and $\mathbf{j}_0(\mathbf{r})$, respectively, must be expressible using the free-space Green tensor (i.e., just as in App. A.1) even if they are regarded as containing medium-response contributions. Alternatively, the same fields must be expressible using the Green tensor that has been constructed from Eq. (2.18), with $\boldsymbol{Q}(\mathbf{r},\mathbf{r}',\omega)$ being the conductivity tensor that accounts precisely for the medium-response contributions. When choosing the latter option, the medium-response contributions have of course to be omitted from $\rho_0(\mathbf{r})$ and $\mathbf{j}_0(\mathbf{r})$, i.e., only the remainders not covered by the conductivity tensor have to be kept explicitly as source terms. Since such a redistribution of sources is possible for an arbitrarily given conductivity tensor $\boldsymbol{Q}(\mathbf{r},\mathbf{r}',\omega)$, and since the choice of $\rho_0(\mathbf{r})$ and $\mathbf{j}_0(\mathbf{r})$ in App. A.1 is also arbitrary, conclusions that are valid for arbitrary Green tensors can be drawn on this basis. Specifically, from (the first line of) the electrostatics formula (A.5), we are permitted to conclude that a finite static (remainder) polarization will properly generate a finite and purely longitudinal electrostatic field only if the leading asymptotic behavior of the Green tensor near $\omega = 0$ is generally given by

$$\boldsymbol{G}(\mathbf{r},\mathbf{r}',\omega) \simeq -\frac{c^2}{\omega^2}\boldsymbol{L}(\mathbf{r},\mathbf{r}'). \tag{A.11}$$

Here, the (non-zero) tensorial coefficient $\boldsymbol{L}(\mathbf{r},\mathbf{r}')$ must be real, longitudinal from the left, and—due to the reciprocity of $\boldsymbol{G}(\mathbf{r},\mathbf{r}',\omega)$—also longitudinal from the right,

$$\int \mathrm{d}^3s\, \boldsymbol{L}(\mathbf{r},\mathbf{s})\cdot\boldsymbol{\Delta}_{\parallel}(\mathbf{s}-\mathbf{r}') = \boldsymbol{L}(\mathbf{r},\mathbf{r}') = \int \mathrm{d}^3s\, \boldsymbol{\Delta}_{\parallel}(\mathbf{r}-\mathbf{s})\cdot\boldsymbol{L}(\mathbf{s},\mathbf{r}'). \tag{A.12}$$

[Note that $\boldsymbol{L}(\mathbf{r},\mathbf{r}')$ is not necessarily equal to $\boldsymbol{\Delta}_{\parallel}(\mathbf{r}-\mathbf{r}')$ in general.] Similarly, from the (first line of the) magnetostatics formula (A.8), we can see that a finite transverse static (remainder) current density will properly generate a finite magnetostatic induction field only if the transverse-from-the-right part $\int \mathrm{d}^3s\, \boldsymbol{G}(\mathbf{r},\mathbf{s},\omega)\cdot\boldsymbol{\Delta}_{\perp}(\mathbf{s}-\mathbf{r}')$ of the Green tensor is generally finite (and in general non-vanishing) at $\omega = 0$. Due to the reciprocity of $\boldsymbol{G}(\mathbf{r},\mathbf{r}',\omega)$, the same is true for the transverse-from-the-left part $\int \mathrm{d}^3s\, \boldsymbol{\Delta}_{\perp}(\mathbf{r}-\mathbf{s})\cdot\boldsymbol{G}(\mathbf{s},\mathbf{r}',\omega)$ of the Green tensor. (In particular, the result of taking the transverse part of the Green tensor from both sides is hence generally finite at $\omega = 0$.) From inspection of Eq. (A.6), we can furthermore see that the finiteness of the transverse-from-the-left part of $\boldsymbol{G}(\mathbf{r},\mathbf{r}',\omega)$ ensures also the absence of a magnetic field in electrostatics, as required. Similarly, we can see from Eq. (A.7) that the finiteness of the transverse-from-the-right part of $\boldsymbol{G}(\mathbf{r},\mathbf{r}',\omega)$ properly ensures also the absence of an electric field in magnetostatics.

From the above, it follows that idealized conductivity tensors that do not show a behavior as required by Eqs. (A.9) and (A.10) will in general give rise to Green tensors that exhibit objectionable properties in the static limit. Such conductivity tensors and Green tensors are thus overly idealized and physically inconsistent, at least with regard to the static limit. Specifically, as Eq. (A.10) cannot be satisfied by any spatially local (isotropic or anisotropic) conductivity tensor, it follows that the concept of a non-vanishing and spatially non-dispersive static conductivity (tensor) suffers from consistency problems. In particular, the permittivity $\varepsilon(\mathbf{r},\omega)$ of a locally responding isotropic dielectric medium—to which corresponds a spatially local conductivity tensor $\boldsymbol{Q}(\mathbf{r},\mathbf{r}',\omega) = -i\omega\varepsilon_0\,[\varepsilon(\mathbf{r},\omega)-1]\,\boldsymbol{I}\,\delta(\mathbf{r}-\mathbf{r}')$—should not be permitted to display a pole at $\omega = 0$ for consistency.

A.3 Proof of the Green-Tensor Integral Relation (2.19)

The linear integro-differential equation (2.18) can be represented as

$$\int \mathrm{d}^3 s\, \boldsymbol{H}(\mathbf{r},\mathbf{s},\omega)\cdot\boldsymbol{G}(\mathbf{s},\mathbf{r}',\omega) = \boldsymbol{I}\delta(\mathbf{r}-\mathbf{r}'), \tag{A.13}$$

where the integral kernel

$$\boldsymbol{H}(\mathbf{r},\mathbf{r}',\omega) = \boldsymbol{\nabla}\times\boldsymbol{\nabla}\times\boldsymbol{I}\delta(\mathbf{r}-\mathbf{r}') - \frac{\omega^2}{c^2}\boldsymbol{I}\delta(\mathbf{r}-\mathbf{r}') - i\mu_0\omega\,\boldsymbol{Q}(\mathbf{r},\mathbf{r}',\omega) \tag{A.14}$$

is reciprocal,

$$\boldsymbol{H}(\mathbf{r},\mathbf{r}',\omega) = \boldsymbol{H}^{\mathsf{T}}(\mathbf{r}',\mathbf{r},\omega), \tag{A.15}$$

since $\boldsymbol{Q}(\mathbf{r},\mathbf{r}',\omega)$ is reciprocal, cf. Eq.(2.9). Hence, the transposed equation of Eq. (A.13) takes the form

$$\int \mathrm{d}^3 s\, \boldsymbol{G}^{\mathsf{T}}(\mathbf{s},\mathbf{r},\omega)\cdot\boldsymbol{H}(\mathbf{s},\mathbf{r}',\omega) = \boldsymbol{I}\delta(\mathbf{r}-\mathbf{r}'). \tag{A.16}$$

Multiplying from the right with $\boldsymbol{G}(\mathbf{r}',\mathbf{s}',\omega)$, integrating over $\mathbf{r}'$, and using Eq. (A.13), one can see that the Green tensor is also reciprocal,

$$\boldsymbol{G}(\mathbf{r},\mathbf{r}',\omega) = \boldsymbol{G}^{\mathsf{T}}(\mathbf{r}',\mathbf{r},\omega). \tag{A.17}$$

Because of Eq. (A.17), the complex conjugate of Eq. (A.16) reads

$$\int \mathrm{d}^3 s\, \boldsymbol{G}^{*}(\mathbf{r},\mathbf{s},\omega)\cdot\boldsymbol{H}^{*}(\mathbf{s},\mathbf{r}',\omega) = \boldsymbol{I}\delta(\mathbf{r}-\mathbf{r}'). \tag{A.18}$$

Taking the dot product of Eq. (A.13) from the left with $\boldsymbol{G}^*(\mathbf{s}',\mathbf{r},\omega)$ and integrating over $\mathbf{r}$, taking the dot product of Eq. (A.18) from the right with $\boldsymbol{G}(\mathbf{r}',\mathbf{s}',\omega)$ and integrating over $\mathbf{r}'$, and subtracting the two resulting equations, one derives

$$\mathrm{Im}\,\boldsymbol{G}(\mathbf{r},\mathbf{r}',\omega) = -\int \mathrm{d}^3s \int \mathrm{d}^3s'\; \boldsymbol{G}(\mathbf{r},\mathbf{s},\omega)\cdot[\mathrm{Im}\,\boldsymbol{H}(\mathbf{s},\mathbf{s}',\omega)]\cdot \boldsymbol{G}^*(\mathbf{s}',\mathbf{r}',\omega). \tag{A.19}$$

From Eq. (A.14) it is seen that

$$\mathrm{Im}\,\boldsymbol{H}(\mathbf{r},\mathbf{r}',\omega) = -\frac{\mathrm{Im}\,\omega^2}{c^2}\boldsymbol{I}\delta(\mathbf{r}-\mathbf{r}') - \mu_0 \mathrm{Re}\,[\omega\boldsymbol{Q}(\mathbf{r},\mathbf{r}',\omega)]. \tag{A.20}$$

Insertion of Eq. (A.20) into Eq. (A.19) and restriction to real frequencies leads, upon recalling Eq. (2.10), to Eq. (2.19).

A.4 Proof of the Fundamental Commutator (2.24)

Using Eqs. (2.20)–(2.23) and recalling the reciprocity of $\boldsymbol{G}(\mathbf{r},\mathbf{r}',\omega)$, we may write

$$\begin{aligned}[\hat{\mathbf{E}}(\mathbf{r}),\hat{\mathbf{B}}(\mathbf{r}')] = -i\mu_0^2 \int_0^\infty \mathrm{d}\omega\,\omega \int_0^\infty \mathrm{d}\omega' \int \mathrm{d}^3s \int \mathrm{d}^3s' \\ \times\Big\{ \boldsymbol{G}(\mathbf{r},\mathbf{s},\omega)\cdot\big[\hat{\underline{\mathbf{j}}}_{\mathrm{N}}(\mathbf{s},\omega),\hat{\underline{\mathbf{j}}}_{\mathrm{N}}^\dagger(\mathbf{s}',\omega')\big]\cdot\boldsymbol{G}^*(\mathbf{s}',\mathbf{r}',\omega') \\ +\boldsymbol{G}^*(\mathbf{r},\mathbf{s},\omega)\cdot\big[\hat{\underline{\mathbf{j}}}_{\mathrm{N}}^\dagger(\mathbf{s},\omega),\hat{\underline{\mathbf{j}}}_{\mathrm{N}}(\mathbf{s}',\omega')\big]\cdot\boldsymbol{G}(\mathbf{s}',\mathbf{r}',\omega')\Big\}\times\overleftarrow{\boldsymbol{\nabla}}'.\end{aligned} \tag{A.21}$$

Applying the commutation relation (2.25) and employing the reality and reciprocity of $\boldsymbol{\sigma}(\mathbf{r},\mathbf{r}',\omega)$ [cf. Eqs. (2.9) and (2.10)], we may carry out one of the frequency integrals to obtain

$$\begin{aligned}[\hat{\mathbf{E}}(\mathbf{r}),\hat{\mathbf{B}}(\mathbf{r}')] = \frac{2\hbar\mu_0^2}{i\pi}\,\mathrm{Re}\int_0^\infty \mathrm{d}\omega\,\omega^2 \\ \times\int \mathrm{d}^3s\int \mathrm{d}^3s'\,\boldsymbol{G}(\mathbf{r},\mathbf{s},\omega)\cdot\boldsymbol{\sigma}(\mathbf{s},\mathbf{s}',\omega)\cdot\boldsymbol{G}^*(\mathbf{s}',\mathbf{r}',\omega)\times\overleftarrow{\boldsymbol{\nabla}}'.\end{aligned} \tag{A.22}$$

By means of the integral relation (2.19), we can now evaluate the spatial integrals. On recalling the relation $\boldsymbol{G}^*(\mathbf{r},\mathbf{r}',\omega) = \boldsymbol{G}(\mathbf{r},\mathbf{r}',-\omega^*)$, we find

$$\begin{aligned}[\hat{\mathbf{E}}(\mathbf{r}),\hat{\mathbf{B}}(\mathbf{r}')] &= \frac{2\hbar\mu_0}{i\pi}\int_0^\infty \mathrm{d}\omega\,\omega\,\mathrm{Im}\,\boldsymbol{G}(\mathbf{r},\mathbf{r}',\omega)\times\overleftarrow{\boldsymbol{\nabla}}' \\ &= \frac{\hbar\mu_0}{i\pi}\left[\int_{-\infty}^\infty \mathrm{d}\omega\,\omega\,\mathrm{Im}\,\boldsymbol{G}(\mathbf{r},\mathbf{r}',\omega)\right]\times\overleftarrow{\boldsymbol{\nabla}}'.\end{aligned} \tag{A.23}$$

Recalling that $\boldsymbol{G}(\mathbf{r},\mathbf{r}',\omega)$ and $\boldsymbol{Q}(\mathbf{r},\mathbf{r}',\omega)$ are (Fourier transformed) response functions, we may conclude from Eq. (2.18) that the leading asymptotic behavior of $\boldsymbol{G}(\mathbf{r},\mathbf{r}',\omega)$ is given by

$$\boldsymbol{G}(\mathbf{r},\mathbf{r}',\omega) \simeq -\frac{c^2}{\omega^2}\boldsymbol{I}\delta(\mathbf{r}-\mathbf{r}') \tag{A.24}$$

along along any direction in the upper ω half-plane (including the real axis). [Alternatively, since within the framework of macroscopic QED the presence of any medium is irrelevant at sufficiently high frequencies, Eq. (A.24) may be derived also from Eqs. (A.2)–(A.4).] Together with the fact that the most singular ($\sim \omega^{-2}$) term of $\boldsymbol{G}(\mathbf{r},\mathbf{r}',\omega)$ at $\omega = 0$ is given by Eq. (A.11), and does not contribute to $\mathrm{Im}\,\boldsymbol{G}(\mathbf{r},\mathbf{r}',\omega)$ for real ω, this shows that the integral within the square brackets in Eq. (A.23) converges. It may be evaluated by contour-integral techniques as

$$\begin{aligned}\int_{-\infty}^{\infty} \mathrm{d}\omega\,\omega\,\mathrm{Im}\,\boldsymbol{G}(\mathbf{r},\mathbf{r}',\omega) &= \mathrm{Im}\,\mathcal{P}\int_{-\infty}^{\infty} \mathrm{d}\omega\,\omega\,\boldsymbol{G}(\mathbf{r},\mathbf{r}',\omega)\\ &= \mathrm{Im}\int_{\mathcal{C}} \mathrm{d}\omega\,\omega\,\boldsymbol{G}(\mathbf{r},\mathbf{r}',\omega) = \pi c^2[\boldsymbol{I}\delta(\mathbf{r}-\mathbf{r}') - \boldsymbol{L}(\mathbf{r},\mathbf{r}')].\end{aligned} \tag{A.25}$$

Here, the principal-value ($\mathcal{P}$) integral has been changed to an integral over a contour $\mathcal{C}$ that consists of an infinitely large semi-circle in the upper half-plane (traversed clockwise), plus an infinitely small semi-circle (traversed counter-clockwise) that avoids the origin in the upper half-plane. Taking into account Eq. (A.24) on the large semi-circle and Eq. (A.11) on the small one, the result given in Eq. (A.25) is obtained. Note that the sub-leading (weaker than ω^{-2}) singular terms of $\boldsymbol{G}(\mathbf{r},\mathbf{r}',\omega)$ at $\omega = 0$ do not contribute, irrespective of the actual nature of the singularity. Inserting Eq. (A.25) in Eq. (A.23), and using Eq. (A.12), we arrive at the desired Eq. (2.24) $[\boldsymbol{\nabla}\times\boldsymbol{I}\delta(\mathbf{r}-\mathbf{r}') = -\boldsymbol{I}\delta(\mathbf{r}-\mathbf{r}')\times\overleftarrow{\boldsymbol{\nabla}}']$.

A.5 Reduced State Space and Super-Selection Rule

Let us consider the state space spanned by the Fock states associated with $\hat{\mathbf{f}}(\mathbf{r},\omega)$ and $\hat{\mathbf{f}}^{\dagger}(\mathbf{r},\omega)$ so that an arbitrary, normalizable state $|\phi\rangle$ in this space can be represented in the form

$$
\begin{aligned}
|\phi\rangle = |0\rangle\langle 0|\phi\rangle &+ \sum_{k_1=1}^{3}\int_0^\infty \mathrm{d}\omega_1 \int \mathrm{d}^3 r_1\, \phi_{k_1}(\mathbf{r}_1,\omega_1)\, |1_{k_1}(\mathbf{r}_1,\omega_1)\rangle \\
&+ \sum_{k_1,k_2=1}^{3}\int_0^\infty \mathrm{d}\omega_1 \int_0^\infty \mathrm{d}\omega_2 \int \mathrm{d}^3 r_1 \int \mathrm{d}^3 r_2 \\
&\times \phi_{k_1 k_2}(\mathbf{r}_1,\omega_1,\mathbf{r}_2,\omega_2)\, |1_{k_1}(\mathbf{r}_1,\omega_1), 1_{k_2}(\mathbf{r}_2,\omega_2)\rangle + \ldots, \qquad \text{(A.26)}
\end{aligned}
$$

where

$$\hat{f}_k(\mathbf{r},\omega)\,|0\rangle = 0, \qquad \text{(A.27)}$$

$$\hat{f}_k^\dagger(\mathbf{r},\omega)\,|0\rangle = |1_k(\mathbf{r},\omega)\rangle\,, \qquad \text{(A.28)}$$

$$\hat{f}_{k_N}^\dagger(\mathbf{r}_N,\omega_N)\cdots\hat{f}_{k_1}^\dagger(\mathbf{r}_1,\omega_1)\,|0\rangle = |1_{k_1}(\mathbf{r}_1,\omega_1),\ldots,1_{k_N}(\mathbf{r}_N,\omega_N)\rangle\,. \qquad \text{(A.29)}$$

The normalization of $|\phi\rangle$ can be obtained by using the formula (which can be viewed as a special case of the Bloch–De Dominicis theorem [34])

$$
\begin{aligned}
&\langle 0|\,\hat{f}_{k_M}(\mathbf{r}_M,\omega_M)\ldots\hat{f}_{k_1}(\mathbf{r}_1,\omega_1)\hat{f}_{k_1'}^\dagger(\mathbf{r}_1',\omega_1')\ldots\hat{f}_{k_N'}^\dagger(\mathbf{r}_N',\omega_N')\,|0\rangle \\
&\qquad = \delta_{MN}\sum_{\pi\in\mathcal{S}_N}\prod_{l=1}^{N}\delta_{k_l,k'_{\pi(l)}}\delta\big(\mathbf{r}_l-\mathbf{r}'_{\pi(l)}\big)\delta\big(\omega_l-\omega'_{\pi(l)}\big) \qquad \text{(A.30)}
\end{aligned}
$$

($\langle 0|0\rangle = 1$; $\mathcal{S}_N$, group of permutations of N objects).

In order to construct a reduced state space in which the operators $\hat{\mathbf{f}}_\lambda(\mathbf{r},\omega)$ and $\hat{\mathbf{f}}_\lambda^\dagger(\mathbf{r},\omega)$ defined by Eq. (2.61) behave like bosonic operators, let us first introduce states $|0\rangle_\lambda$ according to

$$\hat{f}_{\lambda i}(\mathbf{r},\omega)\,|0\rangle_\lambda = 0, \qquad \text{(A.31)}$$

$$\hat{f}_{\lambda i}(\mathbf{r},\omega)\,|0\rangle_{\lambda'} = |0\rangle_{\lambda'}\hat{f}_{\lambda i}(\mathbf{r},\omega) \qquad (\lambda\neq\lambda') \qquad \text{(A.32)}$$

(${}_\lambda\langle 0|0\rangle_\lambda = 1$), such that

$$|0\rangle = \bigotimes_{\lambda=1}^{\Lambda}|0\rangle_\lambda\,. \qquad \text{(A.33)}$$

Now let us introduce, for each λ, an orthogonal projector $\hat{P}_\lambda$ as the sum of orthogonal projectors $\hat{P}_\lambda^{(N)}$,

$$\hat{P}_\lambda = \sum_{N=0}^{\infty}\hat{P}_\lambda^{(N)}, \qquad \text{(A.34)}$$

$$\hat{P}_\lambda^{(N)\dagger} = \hat{P}_\lambda^{(N)}, \qquad \text{(A.35)}$$

$$\hat{P}_\lambda^{(N)}\hat{P}_\lambda^{(N')} = \delta_{NN'}\hat{P}_\lambda^{(N)} \qquad \text{(A.36)}$$

and specify $\hat{P}_\lambda^{(N)}$ in such a way that, when applied to a quantum state of the form (A.26), it picks out the $(N+1)$th term on the right-hand side of Eq. (A.26) and incorporates N position-space projection kernels belonging to the chosen value of λ,

$$\hat{P}_\lambda^{(0)} = |0\rangle_\lambda \langle 0|_\lambda \,, \tag{A.37}$$

$$\begin{aligned}\hat{P}_\lambda^{(N)} = \frac{1}{N!} \sum_{k_1} \int_0^\infty \mathrm{d}\omega_1 \int \mathrm{d}^3 r_1 \sum_{k_2} \int_0^\infty \mathrm{d}\omega_2 \int \mathrm{d}^3 r_2 \cdots \sum_{k_N} \int_0^\infty \mathrm{d}\omega_N \int \mathrm{d}^3 r_N \\ \times \hat{f}^\dagger_{\lambda k_1}(\mathbf{r}_1,\omega_1) \hat{f}^\dagger_{\lambda k_2}(\mathbf{r}_2,\omega_2) \cdots \hat{f}^\dagger_{\lambda k_N}(\mathbf{r}_N,\omega_N) \\ \times \hat{P}_\lambda^{(0)} \hat{f}_{\lambda k_N}(\mathbf{r}_N,\omega_N) \hat{f}_{\lambda k_{N-1}}(\mathbf{r}_{N-1},\omega_{N-1}) \cdots \hat{f}_{\lambda k_1}(\mathbf{r}_1,\omega_1)\end{aligned} \tag{A.38}$$

$(N = 1, 2, \ldots)$. It is not difficult to prove that Eqs. (A.35) and (A.36) are fulfilled, where the latter equation fixes the normalization factor $1/N!$ in Eq. (A.38), and that, in view of Eqs. (A.33) and (A.36), the commutation relation

$$\big[\hat{P}_\lambda^{(N)}, \hat{P}_{\lambda'}^{(N')}\big] = 0 \tag{A.39}$$

holds.

We may now define a reduced state space that contains only those (normalizable) vectors that have the separable form

$$|\phi\rangle^{(\mathrm{red})} = \bigotimes_{\lambda=1}^{\Lambda} |\phi\rangle_\lambda \,, \tag{A.40}$$

$$\hat{P}_\lambda \, |\phi\rangle_\lambda = |\phi\rangle_\lambda \,, \tag{A.41}$$

with each vector $|\phi\rangle_\lambda$ being, by construction, a superposition of vectors

$$\begin{aligned}|N\rangle_\lambda = \sum_{k_1} \int_0^\infty \mathrm{d}\omega_1 \int \mathrm{d}^3 r_1 \ldots \sum_{k_N} \int_0^\infty \mathrm{d}\omega_N \int \mathrm{d}^3 r_N \\ \times C_{\lambda k_1 \ldots \lambda k_N}(\mathbf{r}_1,\omega_1,\cdots,\mathbf{r}_N,\omega_N) \, |1_{\lambda k_1}(\mathbf{r}_1,\omega_1),\ldots,1_{\lambda k_N}(\mathbf{r}_N,\omega_N)\rangle \,,\end{aligned} \tag{A.42}$$

where, in analogy to Eq. (A.29),

$$|1_{\lambda k_1}(\mathbf{r}_1,\omega_1),\ldots,1_{\lambda k_N}(\mathbf{r}_N,\omega_N)\rangle \;=\; \hat{f}^\dagger_{\lambda k_N}(\mathbf{r}_N,\omega_N) \cdots \hat{f}^\dagger_{\lambda k_1}(\mathbf{r}_1,\omega_1) \, |0\rangle_\lambda \,. \tag{A.43}$$

The important feature of these states is that the result of performing the integrations in Eq. (A.42) is not changed if the wave function

$C_{\lambda k_1 \ldots \lambda k_N}(\mathbf{r}_1, \omega_1, \cdots, \mathbf{r}_N, \omega_N)$ is replaced according to

$$C_{\lambda k_1 \ldots \lambda k_N}(\mathbf{r}_1, \omega_1, \cdots, \mathbf{r}_N, \omega_N) \mapsto \int \mathrm{d}^3 r_1' \cdots \int \mathrm{d}^3 r_N' \, (\boldsymbol{P}_\lambda)_{k_1 k_1'}(\mathbf{r}_1, \mathbf{r}_1', \omega_1) \cdots \\ \times (\boldsymbol{P}_\lambda)_{k_N k_N'}(\mathbf{r}_N, \mathbf{r}_N', \omega_N) C_{\lambda k_1' \ldots \lambda k_N'}(\mathbf{r}_1', \omega_1, \cdots, \mathbf{r}_N', \omega_N). \quad \text{(A.44)}$$

It is also not changed if $C_{\lambda k_1 \ldots \lambda k_N}(\mathbf{r}_1, \omega_1, \cdots, \mathbf{r}_N, \omega_N)$ is symmetrized with repect to the labels $1, \ldots, N$. Wave functions that can be reduced to the same standardized wave function by these operations are thus fully equivalent representatives of the same vector. Without loss of generality, one can thus adopt the convention to employ only such standardized wave functions.

The commutation relation (2.62) implies that

$$e^{\epsilon \hat{f}_{\lambda k}(\mathbf{r},\omega)} \hat{f}^\dagger_{\lambda' k'}(\mathbf{r}', \omega') e^{-\epsilon \hat{f}_{\lambda k}(\mathbf{r},\omega)} = \hat{f}^\dagger_{\lambda' k'}(\mathbf{r}', \omega') + \epsilon \delta_{\lambda\lambda'} (\boldsymbol{P}_\lambda)_{kk'}(\mathbf{r}, \mathbf{r}', \omega) \delta(\omega - \omega') \quad \text{(A.45)}$$

with ϵ being a parameter. As Eq. (A.45) is a similarity transformation, it generalizes to

$$e^{\epsilon \hat{f}_{\lambda k}(\mathbf{r},\omega)} F[\hat{f}^\dagger_{\lambda' k'}(\mathbf{r}', \omega')] e^{-\epsilon \hat{f}_{\lambda k}(\mathbf{r},\omega)} \\ = F[\hat{f}^\dagger_{\lambda' k'}(\mathbf{r}', \omega') + \epsilon \delta_{\lambda\lambda'} (\boldsymbol{P}_\lambda)_{kk'}(\mathbf{r}, \mathbf{r}', \omega) \delta(\omega - \omega')] \quad \text{(A.46)}$$

where $F = F[\hat{f}^\dagger_{\lambda' k'}(\mathbf{r}', \omega')]$ is any well-behaved functional of $\hat{f}^\dagger_{\lambda' k'}(\mathbf{r}', \omega')$. Comparison of the terms of first order in ϵ on both sides yields

$$\left[\hat{f}_{\lambda k}(\mathbf{r}, \omega), F[\hat{f}^\dagger_{\lambda' k'}(\mathbf{r}', \omega')]\right] \\ = \left\{ \frac{\partial}{\partial \epsilon} F[\hat{f}^\dagger_{\lambda' k'}(\mathbf{r}', \omega') + \epsilon \delta_{\lambda\lambda'} (\boldsymbol{P}_\lambda)_{kk'}(\mathbf{r}, \mathbf{r}', \omega) \delta(\omega - \omega')] \right\}_{\epsilon=0}. \quad \text{(A.47)}$$

Let us consider the particular functional $F_N[\hat{f}^\dagger_{\lambda' k'}(\mathbf{r}', \omega')]$ appearing in Eqs. (A.42) and (A.43),

$$F_N[\hat{f}^\dagger_{\lambda' k'}(\mathbf{r}', \omega')] = \sum_{k_1} \int_0^\infty \mathrm{d}\omega_1 \int \mathrm{d}^3 r_1 \ldots \sum_{k_N} \int_0^\infty \mathrm{d}\omega_N \int \mathrm{d}^3 r_N \\ \times C_{\lambda k_1 \ldots \lambda k_N}(\mathbf{r}_1, \omega_1, \cdots, \mathbf{r}_N, \omega_N) \hat{f}^\dagger_{\lambda k_N}(\mathbf{r}_N, \omega_N) \cdots \hat{f}^\dagger_{\lambda k_1}(\mathbf{r}_1, \omega_1). \quad \text{(A.48)}$$

If the convention to use only standardized wave functions is adopted, one may write

$$F_N[\hat{f}^\dagger_{\lambda' k'}(\mathbf{r}', \omega') + \epsilon \delta_{\lambda\lambda'} (\boldsymbol{P}_\lambda)_{kk'}(\mathbf{r}, \mathbf{r}', \omega') \delta(\omega - \omega')] \\ = F_N[\hat{f}^\dagger_{\lambda' k'}(\mathbf{r}', \omega') + \epsilon \delta_{\lambda\lambda'} \delta_{kk'} \delta(\mathbf{r} - \mathbf{r}') \delta(\omega - \omega')], \quad \text{(A.49)}$$

which means that the right-hand side of Eq. (A.47) may be evaluated, for this functional, just as an ordinary functional derivative, i.e.,

$$\Big[\hat{f}_{\lambda k}(\mathbf{r},\omega), F_N[\hat{f}^\dagger_{\lambda' k'}(\mathbf{r}',\omega')]\Big] = \frac{\delta F_N[\hat{f}^\dagger_{\lambda' k'}(\mathbf{r}',\omega')]}{\delta \hat{f}^\dagger_{\lambda k}(\mathbf{r},\omega)}. \tag{A.50}$$

But since, due to the definition of the reduced state space, only commutators of the type (A.50) (for all N) are required, and since Eq. (A.50) can be obtained from Eq. (2.69) in the same way that Eq. (A.47) has been obtained from Eq. (2.62), Eq. (2.69) is generally valid for the reduced state space.

A.6 Proof of Eqs. (3.17)–(3.25)

From Eqs. (2.22), (2.23), (3.44), and (3.45) together with the commutation relation (2.25) we derive, on recalling the reciprocity of the Green tensor and the relation (2.19),

$$\begin{aligned}\big[\underline{\hat{\rho}}(\mathbf{r},\omega), \underline{\hat{\mathbf{E}}}^\dagger(\mathbf{r}',\omega')\big] &= \frac{\hbar}{\pi}\frac{\omega^2}{c^2}\,\delta(\omega-\omega')\boldsymbol{\nabla}\cdot\mathrm{Im}\,\boldsymbol{G}(\mathbf{r},\mathbf{r}',\omega)\\ &= -\big[\underline{\hat{\rho}}^\dagger(\mathbf{r},\omega), \underline{\hat{\mathbf{E}}}(\mathbf{r}',\omega')\big],\end{aligned} \tag{A.51}$$

$$\begin{aligned}\big[\underline{\hat{\mathbf{j}}}(\mathbf{r},\omega), \underline{\hat{\mathbf{B}}}^\dagger(\mathbf{r}',\omega')\big] &= -\frac{\hbar}{\pi}\,\delta(\omega-\omega')\bigg[\bigg(\boldsymbol{\nabla}\times\boldsymbol{\nabla}\times-\frac{\omega^2}{c^2}\bigg)\mathrm{Im}\,\boldsymbol{G}(\mathbf{r},\mathbf{r}',\omega)\times\overleftarrow{\boldsymbol{\nabla}}'\bigg]\\ &= -\big[\underline{\hat{\mathbf{j}}}^\dagger(\mathbf{r},\omega), \underline{\hat{\mathbf{B}}}(\mathbf{r}',\omega')\big],\end{aligned} \tag{A.52}$$

$$\begin{aligned}\big[\underline{\hat{\rho}}(\mathbf{r},\omega), \underline{\hat{\mathbf{B}}}^\dagger(\mathbf{r}',\omega')\big] &= -\frac{\hbar}{\pi}\frac{i\omega}{c^2}\,\delta(\omega-\omega')\boldsymbol{\nabla}\cdot\mathrm{Im}\,\boldsymbol{G}(\mathbf{r},\mathbf{r}',\omega)\times\overleftarrow{\boldsymbol{\nabla}}'\\ &= \big[\underline{\hat{\rho}}^\dagger(\mathbf{r},\omega), \underline{\hat{\mathbf{B}}}(\mathbf{r}',\omega')\big],\end{aligned} \tag{A.53}$$

and

$$\begin{aligned}\big[\underline{\hat{\mathbf{j}}}(\mathbf{r},\omega), \underline{\hat{\mathbf{E}}}(\mathbf{r}',\omega')^\dagger\big] &= -\frac{\hbar}{\pi}\,i\omega\,\delta(\omega-\omega')\bigg[\bigg(\boldsymbol{\nabla}\times\boldsymbol{\nabla}\times-\frac{\omega^2}{c^2}\bigg)\mathrm{Im}\,\boldsymbol{G}(\mathbf{r},\mathbf{r}',\omega)\bigg]\\ &= \big[\underline{\hat{\mathbf{j}}}^\dagger(\mathbf{r},\omega), \underline{\hat{\mathbf{E}}}(\mathbf{r}',\omega')\big].\end{aligned} \tag{A.54}$$

Equations (A.51) and (A.52) obviously imply the commutation relations

$$\begin{aligned}\big[\hat{\rho}(\mathbf{r}), \hat{\mathbf{E}}(\mathbf{r}')\big] = \int_0^\infty \mathrm{d}\omega \int_0^\infty \mathrm{d}\omega' \bigg\{ &\big[\underline{\hat{\rho}}(\mathbf{r},\omega), \underline{\hat{\mathbf{E}}}^\dagger(\mathbf{r}',\omega')\big]\\ &+ \big[\underline{\hat{\rho}}^\dagger(\mathbf{r},\omega), \underline{\hat{\mathbf{E}}}(\mathbf{r}',\omega')\big]\bigg\} = 0\end{aligned} \tag{A.55}$$

and

$$\left[\hat{\mathbf{j}}(\mathbf{r}),\hat{\mathbf{B}}(\mathbf{r}')\right] = \int_0^\infty \mathrm{d}\omega \int_0^\infty \mathrm{d}\omega' \left\{ \left[\underline{\hat{\mathbf{j}}}(\mathbf{r},\omega),\underline{\hat{\mathbf{B}}}^\dagger(\mathbf{r}',\omega')\right] + \left[\underline{\hat{\mathbf{j}}}^\dagger(\mathbf{r},\omega),\underline{\hat{\mathbf{B}}}(\mathbf{r}',\omega')\right] \right\} = 0, \quad \text{(A.56)}$$

and hence Eqs. (3.17) and (3.19) are seen to hold. Note in particular that the commutation relation $\left[\hat{\rho}(\mathbf{r}),\hat{\mathbf{E}}^\perp(\mathbf{r}')\right]=0$ is valid. From Eqs. (A.53) and (A.54), respectively, it follows that

$$\left[\hat{\rho}(\mathbf{r}),\hat{\mathbf{B}}(\mathbf{r}')\right] = -\frac{2i\hbar}{\pi c^2}\int_0^\infty \mathrm{d}\omega\,\omega\boldsymbol{\nabla}\cdot \mathrm{Im}\,\boldsymbol{G}(\mathbf{r},\mathbf{r}',\omega)\times\overleftarrow{\boldsymbol{\nabla}}' \qquad \text{(A.57)}$$

and

$$\left[\hat{\mathbf{j}}(\mathbf{r}),\hat{\mathbf{E}}(\mathbf{r}')\right] = -\frac{2i\hbar}{\pi}\int_0^\infty \mathrm{d}\omega\,\omega\left[\left(\boldsymbol{\nabla}\times\boldsymbol{\nabla}\times-\frac{\omega^2}{c^2}\right)\mathrm{Im}\,\boldsymbol{G}(\mathbf{r},\mathbf{r}',\omega)\right]. \qquad \text{(A.58)}$$

To further evaluate the integrals in Eqs. (A.57) and (A.58), we recall that both the conductivity tensor $\boldsymbol{Q}(\mathbf{r},\mathbf{r}',\omega)$ and the Green tensor $\boldsymbol{G}(\mathbf{r},\mathbf{r}',\omega)$ are (Fourier-transformed) response functions and have an asymptotic behavior for large ω in the upper half-plane as specified by Eqs. (3.21) and (A.24), respectively. Recalling that $\boldsymbol{G}^*(\mathbf{r},\mathbf{r}',\omega) = \boldsymbol{G}(\mathbf{r},\mathbf{r}',-\omega^*)$, and making use of Eq. (A.25), we may easily evaluate the integral in Eq. (A.57) to prove Eq. (3.18),

$$\left[\hat{\rho}(\mathbf{r}),\hat{\mathbf{B}}(\mathbf{r}')\right] = -\frac{i\hbar}{\pi c^2}\boldsymbol{\nabla}\cdot\left[\int_{-\infty}^\infty \mathrm{d}\omega\,\omega\,\mathrm{Im}\,\boldsymbol{G}(\mathbf{r},\mathbf{r}',\omega)\right]\times\overleftarrow{\boldsymbol{\nabla}}' = 0. \qquad \text{(A.59)}$$

To evaluate Eq. (A.58), we take into account that according to Eq. (2.18) the relation

$$\left(\boldsymbol{\nabla}\times\boldsymbol{\nabla}\times-\frac{\omega^2}{c^2}\right)\mathrm{Im}\,\boldsymbol{G}(\mathbf{r},\mathbf{r}',\omega) = \mu_0\,\mathrm{Re}\left[\omega\int \mathrm{d}^3s\,\boldsymbol{Q}(\mathbf{r},\mathbf{s},\omega)\cdot\boldsymbol{G}(\mathbf{s},\mathbf{r}',\omega)\right] \qquad \text{(A.60)}$$

may be used on the real ω axis. Inserting this relation into Eq. (A.58), we see that the evaluation of Eq. (A.58) can be done in very much the same way as the evaluation of Eq. (A.57). Noting that there is no problem at $\omega=0$ due to Eqs.(A.9) and (A.11) and making use of Eq. (A.24) and (3.21), we derive

$$\begin{aligned}\left[\hat{\mathbf{j}}(\mathbf{r}),\hat{\mathbf{E}}(\mathbf{r}')\right] &= -\frac{i\hbar\mu_0}{\pi}\mathrm{Re}\int_{-\infty}^\infty \mathrm{d}\omega\,\omega^2\int \mathrm{d}^3s\,\boldsymbol{Q}(\mathbf{r},\mathbf{s},\omega)\cdot\boldsymbol{G}(\mathbf{s},\mathbf{r}',\omega)\\ &= \frac{i\hbar}{\varepsilon_0}\mathrm{Im}\,\boldsymbol{Q}^{(-1)}(\mathbf{r},\mathbf{r}'),\end{aligned} \qquad \text{(A.61)}$$

which is just Eq. (3.25).

For a consistency check of the commutation relation (A.61), let us consider a set of atoms, with each of them having one valence electron (e, charge; m, mass). Let $\mathbf{r}_A$ be the (fixed) positions and $\hat{\mathbf{s}}_A$ the relative coordinates of the electrons. The microscopic (electron) current density is then given by

$$\hat{\mathbf{j}}(\mathbf{r}) = e\sum_A \dot{\hat{\mathbf{s}}}_A \delta(\mathbf{r}-\mathbf{r}_A-\hat{\mathbf{s}}_A). \tag{A.62}$$

By assuming minimal coupling and Coulomb gauge, the canonical momenta of the electrons commute with the vector potential $\hat{\mathbf{A}}(\mathbf{r})$, whose conjugate momentum field is $-\varepsilon_0\hat{\mathbf{E}}^{\perp}(\mathbf{r})$. Hence, we derive

$$\begin{aligned}\left[\hat{\mathbf{j}}(\mathbf{r}),\hat{\mathbf{E}}^{\perp}(\mathbf{r}')\right] &= -\frac{e^2}{m}\sum_A \delta(\mathbf{r}-\mathbf{r}_A-\hat{\mathbf{s}}_A)\left[\hat{\mathbf{A}}(\mathbf{r}_A+\hat{\mathbf{s}}_A),\hat{\mathbf{E}}^{\perp}(\mathbf{r}')\right]\\ &= \frac{i\hbar}{\varepsilon_0}\frac{e^2}{m}\sum_A \delta(\mathbf{r}-\mathbf{r}_A-\hat{\mathbf{s}}_A)\boldsymbol{\Delta}_{\perp}(\mathbf{r}_A+\hat{\mathbf{s}}_A-\mathbf{r}')\\ &= \frac{i\hbar}{\varepsilon_0}\frac{e^2}{m}\sum_A \delta(\mathbf{r}-\mathbf{r}_A-\hat{\mathbf{s}}_A)\boldsymbol{\Delta}_{\perp}(\mathbf{r}-\mathbf{r}'). \end{aligned}\tag{A.63}$$

In the macroscopic theory, the sum of the δ-functions in Eq. (A.63) is expected to be replaced according to

$$\sum_A \delta(\mathbf{r}-\mathbf{r}_A-\hat{\mathbf{s}}_A) \mapsto \sum_A d(\mathbf{r}-\mathbf{r}_A-\hat{\mathbf{s}}_A), \tag{A.64}$$

where $d(\mathbf{r})$ is a well-behaved function with unit integral, $\int \mathrm{d}^3r\, d(\mathbf{r})=1$. Further, in order to produce reasonable coarse-graining, $d(\mathbf{r})$ must be sufficiently flat in the sense that the change of $d(\mathbf{r})$ on atomic length scales can be regarded as being negligibly small. With the $\hat{\mathbf{s}}_A$ acting on well localized electronic bound states, we may hence write $d(\mathbf{r}-\mathbf{r}_A-\hat{\mathbf{s}}_A)\simeq d(\mathbf{r}-\mathbf{r}_A)$. Thus,

$$\sum_A d(\mathbf{r}-\mathbf{r}_A-\hat{\mathbf{s}}_A) \simeq \sum_A d(\mathbf{r}-\mathbf{r}_A) = \eta(\mathbf{r}), \tag{A.65}$$

where $\eta(\mathbf{r})$ is the number density $\eta(\mathbf{r})$ of the atoms, and the macroscopic version of Eq. (A.63) reads

$$\left[\hat{\mathbf{j}}(\mathbf{r}),\hat{\mathbf{E}}^{\perp}(\mathbf{r}')\right] = \frac{i\hbar}{\varepsilon_0}\frac{e^2}{m}\eta(\mathbf{r})\boldsymbol{\Delta}_{\perp}(\mathbf{r}-\mathbf{r}'). \tag{A.66}$$

Recalling that for a locally responding (magneto-)dielectric medium the macroscopically derived Eq. (A.61) leads, in particular, to Eq. (3.25), we observe that from a comparison of Eq. (A.66) with Eq. (3.25) the relation

$$\Omega_\varepsilon^2(\mathbf{r}) = \frac{e^2 \eta(\mathbf{r})}{\varepsilon_0 m} \tag{A.67}$$

is suggested to be valid. In Ref. [R3], Eq. (A.67) has in fact been shown to follow from a harmonic-oscillator model of a locally responding dielectric.

Publications and Talks

[R1] C. Raabe, L. Knöll, and D.-G. Welsch, *Three-dimensional Casimir force between absorbing multilayer dielectrics*, Phys. Rev. A **68** 033810 (2003).

[R2] C. Raabe, L. Knöll, and D.-G. Welsch, *Casimir stress on lossy magnetodielectric spheres*, `arXiv:quant-ph/0309179` (2003).

[R3] C. Raabe and D.-G. Welsch, *Casimir force acting on magnetodielectric bodies embedded in media*, Phys. Rev. A **71** 013814 (2005).

[R4] C. Raabe and D.-G. Welsch, *Matter-screened Casimir force and Casimir-Polder force in planar structures*, J. Opt. B: Quantum Semiclass. Opt. **7** S610 (2005).

[R5] C. Raabe and D.-G. Welsch, *Reply to* "Comment on *'Casimir force acting on magnetodielectric bodies embedded in media'*", Phys. Rev. A **73** 047802 (2006).

[R6] C. Raabe and D.-G. Welsch, *Dispersive forces on bodies and atoms: A unified approach*, Phys. Rev. A **73** 063822 (2006).

[R7] C. Raabe and D.-G. Welsch, *Dispersion Forces within the Framework of Macroscopic QED*, Acta Phys. Hung. A **26** 3 (2006).

[R8] C. Raabe and D.-G. Welsch, *Unified approach to dispersion forces within macroscopic QED*, Hyperfine Interac. **172** 149 (2006).

[R9] C. Raabe and D.-G. Welsch, *Lorentz-force approach to the Casimir force*, J. Phys.: Conf. Ser. **36** 159 (2006).

[R10] C. Raabe, S. Scheel, and D.-G. Welsch, *Unified approach to QED in arbitrary linear media*, Phys. Rev. A **75** 053813 (2007).

[R11] C. Raabe and D.-G. Welsch, *QED in arbitrary linear media: amplifying media*, EPJST **160** 371 (2008).

[R12] C. Raabe, *Casimir force for realistic materials*, talk given at Imperial College London (11/2003).

[R13] C. Raabe and D.-G. Welsch, *Casimir force acting on bodies in media: Why textbooks are wrong*, talk given at ICSSUR 2005 conference (05/2005).

Bibliography

[1] E. Buks and M. L. Roukes, Phys. Rev. B **63**, 033402 (2001).

[2] F. W. Delrio, M. P. de Boer, J. A. Knapp, E. D. Reedy, Jr., P. J. Clews, and M. L. Dunn, Nat. Mater. **4**, 629 (2005).

[3] S.-A. Zhou, Int. J. Eng. Sci. **41**, 313 (2003).

[4] A. Ashourvan, M. Miri, and R. Golestanian, Phys. Rev. Lett. **98**, 140801 (2007).

[5] P. W. Milonni, *The Quantum Vacuum—An Introduction to Quantum Electrodynamics* (Academic Press, Inc., San Diego, 1994).

[6] F. London, Z. Phys. **63**, 245 (1930).

[7] F. London, Trans. Faraday Soc. **33**, 8 (1937).

[8] H. B. G. Casimir and D. Polder, Phys. Rev. **73**, 360 (1948).

[9] M. Bordag, U. Mohideen, and V. M. Mostepanenko, Phys. Rep. **353**, 1 (2001).

[10] E. M. Lifshitz, Zh. Eksp. Teor. Fiz. **29**, 94 (1955).

[11] J. Schwinger, L. L. DeRaad, Jr., and K. A. Milton, Ann. Phys. (N.Y.) **115**, 1 (1978).

[12] M. S. Tomaš, Phys. Rev. A **71**, 060101(R) (2005).

[13] M. S. Tomaš, Phys. Rev. A **72**, 034104 (2005). Note erratum Phys. Rev. A **73**, 039904(E) (2006).

[14] M. S. Tomaš, Fiz. A **14**, 29 (2005).

[15] A. A. Abrikosov, L. P. Gorkov, and I. E. Dzyaloshinski, *Methods of Quantum Field Theory in Statistical Physics* (Dover Publications, Inc., New York, 1975).

[16] K. A. Milton and Y. J. Ng, Phys. Rev. E **57**, 5504 (1998).

[17] I. Brevik, V. N. Marachevsky, and K. A. Milton, Phys. Rev. Lett. **82**, 3948 (1999).

[18] G. Barton, J. Phys. A Math. Gen. **32**, 525 (1999).

[19] M. J. Renne, Physica **56**, 125 (1971).

[20] P. W. Milonni and P. B. Lerner, Phys. Rev. A **46**, 1185 (1992).

[21] S. Y. Buhmann and D.-G. Welsch, Appl. Phys. B **82**, 189 (2006).

[22] V. L. Ginzburg, *Applications of Electrodynamics in Theoretical Physics and Astrophysics* (Gordon and Breach, New York, 1989), 2nd rev. edn.

[23] M. S. Tomaš, Phys. Rev. A **66**, 052103 (2002).

[24] F. Zhou and L. Spruch, Phys. Rev. A **52**, 297 (1995).

[25] O. Kenneth and I. Klich, Phys. Rev. Lett. **97**, 160401 (2006).

[26] J. L. Birman and J. J. Sein, Phys. Rev. B **6**, 2482 (1972).

[27] A. A. Maradudin and D. L. Mills, Phys. Rev. B **7**, 2787 (1973).

[28] G. S. Agarwal, D. N. Pattanayak, and E. Wolf, Phys. Rev. Lett. **27**, 1022 (1971).

[29] G. S. Agarwal, D. N. Pattanayak, and E. Wolf, Phys. Rev. B **10**, 1447 (1974).

[30] G. S. Agarwal, D. N. Pattanayak, and E. Wolf, Phys. Rev. B **11**, 1342 (1975).

[31] D. B. Melrose and R. C. McPhedran, *Electromagnetic processes in dispersive media* (Cambridge University Press, Cambridge, 1991).

[32] S. Savasta, O. Di Stefano, and R. Girlanda, Phys. Rev. A **65**, 043801 (2002).

[33] S. Savasta, O. Di Stefano, and R. Girlanda, J. Opt. A: Pure. Appl. Opt. **4**, S95 (2002).

[34] R. Kubo, M. Toda, and N. Hashitsume, *Statistical Physics II: Nonequilibrium Statistical Mechanics* (Springer-Verlag, Berlin, 1991), 2nd edn.

[35] L. D. Landau and E. M. Lifschitz, *Lehrbuch der theoretischen Physik Bd. 5—Statistische Physik* (Akademie Verlag, Berlin, 1966).

[36] H. M. Nussenzveig, *Causality and Dispersion Relations* (Academic Press, London, 1972).

[37] D. B. Melrose and R. J. Stoneham, J. Phys. A: Math. Gen. **10**, L17 (1977).

[38] C. W. Gardiner, *Handbook of Stochastic Methods for Physics, Chemistry and the Natural Sciences* (Springer-Verlag, Berlin, 1990), 2nd edn.

[39] C. W. Gardiner and P. Zoller, *Quantum Noise* (Springer, Berlin, 2004), 3rd edn.

[40] G. W. Ford, J. T. Lewis, and R. F. O'Connell, Phys. Rev. A **37**, 4419 (1988).

[41] M. Lax, Phys. Rev. **145**, 110 (1966).

[42] W. Vogel and D.-G. Welsch, *Quantum Optics* (Wiley-VCH, Weinheim, 2006), 3rd edn.

[43] L. Knöll, S. Scheel, and D.-G. Welsch, in *Coherence and Statistics of Photons and Atoms*, chap. 1 (Wiley, New York, 2001).

[44] S. Scheel, L. Knöll, and D.-G. Welsch, Phys. Rev. A **58**, 700 (1998).

[45] T. D. Ho, S. Y. Buhmann, L. Knöll, D.-G. Welsch, S. Scheel, and J. Kästel, Phys. Rev. A **68**, 043816 (2003).

[46] O. Di Stefano, S. Savasta, and R. Girlanda, J. Opt. B: Quantum Semiclass. Opt. **3**, 288 (2001).

[47] M. Born and E. Wolf, *Priciples of Optics* (Cambridge University Press, Cambridge, United Kingdom, 1998), 6th (corrected) edn.

[48] W. C. Chew, *Waves and Fields in Inhomogeneous Media*, IEEE Press Series on Electromagnetic Waves (IEEE Press, New York, 1995).

[49] L. D. Landau and E. M. Lifschitz, *Lehrbuch der Theoretischen Physik Bd. 8—Elektrodynamik der Kontinua* (Akademie Verlag, Berlin, 1967).

[50] U. Leonhardt and T. G. Philbin, New J. Phys. **9**, 254 (2007).

[51] B. Huttner and S. M. Barnett, Phys. Rev. A **46**, 4306 (1992).

[52] L. G. Suttorp and M. Wubs, Phys. Rev. A **70**, 013816 (2004).

[53] L. G. Suttorp, J. Phys. A: Math. Theor. **40**, 3697 (2007).

[54] A. D. McLachlan, Proc. R. Soc. A **271**, 387 (1963). Mol. Phys. **7** 381.

[55] G. S. Agarwal, Phys. Rev. A **11**, 243 (1975).

[56] J. M. Wylie and J. E. Sipe, Phys. Rev. A **30**, 1185 (1984).

[57] C. Henkel, K. Joulain, J. P. Mulet, and J. J. Greffet, J. Opt. A: Pure Appl. Opt. **4**, 109 (2002).

[58] S. Y. Buhmann, T. D. Ho, and D.-G. Welsch, J. Opt B: Quantum Semiclass. Opt. **6**, S127 (2004).

[59] J. Mahanty and B. W. Ninham, *Dispersion Forces* (Academic Press, London, 1976).

[60] Y. N. Obukhov and F. W. Hehl, Phys. Lett. A **311**, 277 (2003).

[61] G. B. Walker and G. Walker, Can. J. Phys. **55**, 2121 (1977).

[62] G. B. Walker, D. G. Lahoz, and G. Walker, Can. J. Phys. **53**, 2577 (1975).

[63] G. B. Walker and D. G. Lahoz, Nature (London) **253**, 339 (1975).

[64] G. B. Walker and G. Walker, Nature (London) **263**, 401 (1976).

[65] G. B. Walker and G. Walker, Nature (London) **265**, 324 (1977).

[66] R. P. James, Proc. Natl. Acad. Sci. USA **61**, 1149 (1968).

[67] R. P. James, *Force on permeable matter in time-varying fields*, Ph.D. thesis, Department of Electrical Engineering, Stanford University (1968).

[68] I. Brevik, Phys. Rep. **52**, 133 (1979).

[69] D. F. Nelson, Phys. Rev. A **44**, 3985 (1991).

[70] P. Poincelot, C. R. Seances Acad. Sci. Ser.B **264**, 1064, 1179, 1225, 1560 (1967).

[71] G. H. Livens, *The Theory of Electricity* (Cambridge University Press, Cambridge, England, 1926).

[72] G. S. Agarwal and E. Wolf, Phys. Rev. D **2**, 2187 (1970).

[73] G. L. Klimchitskaya and V. M. Mostepanenko, Phys. Rev. A **63**, 062108 (2001).

[74] M. Bordag, B. Geyer, G. L. Klimchitskaya, and V. M. Mostepanenko, Phys. Rev. Lett. **85**, 503 (2000).

[75] B. E. Sernelius and M. Boström, Phys. Rev. Lett. **87**, 259101 (2001).

[76] M. Bordag, B. Geyer, G. L. Klimchitskaya, and V. M. Mostepanenko, Phys. Rev. Lett. **87**, 259102 (2001).

[77] L. P. Pitaevskii, Phys. Rev. A. **73** (2006).

[78] J. D. Jackson, *Classical Electrodynamics* (Wiley, New York, 1999), 3rd edn.

[79] M. S. Tomaš, Phys. Rev. A **51**, 2545 (1995).

[80] J. Mahanty and B. W. Ninham, J. Phys. A **5**, 1447 (1972).

[81] J. Mahanty and B. W. Ninham, J. Phys. A **6**, 1140 (1973).

[82] M. Marcovitch and H. Diamant, Phys. Rev. Lett. **95**, 223203 (2005).

[83] H. B. G. Casimir, Proc. K. Ned. Akad. Wet. **51**, 793 (1948).

[84] M. Schaden, L. Spruch, and F. Zhou, Phys. Rev. A **57**, 1108 (1998).

Zusammenfassung

In dieser Arbeit wird ein sehr allgemein gültiges Quantisierungsschema für das makroskopische elektromagnetische Feld in beliebigen linear reagierenden (ruhenden) Medien dargestellt. Es bietet einen einheitlichen Zugang zur QED in solchen Medien. Durch die Charakterisierung der Medien mittels eines im allgemeinen nichtlokalen Leitfähigkeitstensors können alle denkbaren (makroskopischen) Medieneigenschaften erfaßt werden, speziell auch räumliche Dispersion. Zentrale Größen der Theorie sind die Rauschstromdichte, die mit der im Gleichgewicht unweigerlich auftretenden Absorption einhergeht, die der Rauschstromdichte zugeordneten bosonischen dynamischen Variablen und der Greentensor der makroskopischen Maxwellgleichungen, in den die Medieneigenschaften indirekt über den Leitfähigkeitstensor einfließen. Eventuell zusätzlich zu einem Medium vorhandene geladene Teilchen können ohne Schwierigkeiten anhand der üblichen Verfahrensweisen angekoppelt werden, wobei in diesem Zusammenhang das mediengestützte (bereits mit dem Hintergrundmedium wechselwirkende) Feld die Rolle des freien Feldes spielt. Obwohl linear verstärkende Medien strenggenommen den üblichen Rahmen der linearen Response-Theorie sprengen, kann das Quantisierungsschema auch auf sie angewandt werden, wobei aber einige Änderungen nötig werden.
Eine sorgfältige Analyse der dynamischen Variablen und möglicher quasilokaler Näherungsausdrücke für den Leitfähigkeitstensor zeigt, dass schon früher angegebene Quantisierungsschemata für lokal reagierende Medien Spezialfälle des allgemeinen Schemas sind. Speziell kann ein lokal reagierendes Magnetodielektrikum als quasilokaler Grenzfall eines isotropen, räumlich dispersiven Mediums ohne optische Aktivität aufgefaßt werden, wobei die lokale dielektrische Funktion und die lokale magnetische Permeabilität lediglich zwei Anteile ein und desselben quasi-lokalen Leitfähigkeitstensors bilden. Die Anwendung der allgemeinen Theorie zeigt

dann, dass zur Quantisierung des Feldes in einem solchen Medium nur ein einziger Satz fundamentaler Bose-Variablen erforderlich ist. Die Benutzung nur dieses einen Satzes besagt allgemein gesprochen, dass die in die Maxwellgleichungen einfließende Rauschstromdichte nicht in verschiedene (etwa Polarisations- und Magnetisierungs-)Anteile zerlegt wird, die als unabhängig und verschiedenen Freiheitsgraden zugehörig betrachtet werden, sondern sie im Gegenteil als eine Einheit behandelt wird. Dieser Standpunkt könnte sich beispielsweise im Hinblick auf die Behandlung bewegter Medien als vorteilhaft erweisen, da er gewiß die Transformation in verschiedene Bezugssysteme vereinfacht. Alternativ ist aber auch die Einführung zweier (bzw. mehrerer) unabhängiger Sätze fundamentaler Bose-Variablen möglich, wodurch aber gewisse Wechselwirkungen (durch Superauswahlregeln) aus der Theorie von vornherein ausgeschlossen werden.
Das allgemeine Quantisierungsschema wurde dann angewendet, um das Problem der Dispersionskräfte zu behandeln. Indem selbige als rein elektromagnetische Kräfte aufgefaßt werden – nämlich als die Lorentzkräfte, die das fluktuierende elektromagnetische Vakuumfeld auf diejenigen Ladungen und Ströme ausübt, die aus Sicht der makroskopischen linearen Elektrodynamik einen materiellen Körper ausmachen – gelingt auch hier eine konzeptionelle Vereinfachung. Casimir-, Casimir–Polder- und van der Waals-Kräfte werden so auf eine einheitliche theoretische Grundlage gestellt, die ihrer gemeinsamen physikalischen Ursache gerecht wird. Die Dispersionskräfte auf Makroobjekte, Mikroobjekte und sogar einzelne Atome (im Grundzustand) können somit in einheitlicher Weise berechnet werden. Aufgrund der Auffassung als rein elektromagnetische Kräfte sind sonstige Kräfte, die in der Praxis eine Kompensation der eigentlichen Dispersionskräfte bewirken können, in den Rechnungen freilich nicht enthalten. Umgekehrt liefert aber die Theorie so natürlich genau die zur Kompensation erforderliche Größe solcher Zusatzkräfte. Im Gegensatz zu dieser Betrachtungsweise versuchen Theorien, die (anstelle des Maxwell'schen) mit dem Minkowski'schen Spannungstensor oder verwandten Größen operieren, zusätzliche kompensierende Kraftanteile, die zu den eigentlichen Dispersionskräften hinzutreten können, den letzteren von vornherein hinzuzuschlagen, wobei aber Widersprüche in Erscheinung treten. Es ist wahrscheinlich, dass sich ein solcher Ansatz ohne detaillierte Modellannahmen über die konkret vorliegenden mikroskopischen Eigenschaften der Körper prinzipiell nicht konsistent umsetzen läßt.
Im Rahmen der genannten Auffassung als Lorentzkräfte wurden allgemein-

gültige Formeln für die Dispersionskräfte, die auf Körper oder Teile von ihnen wirken, hergeleitet. Sie sind auf beliebige räumliche Konfigurationen und beliebige linear reagierende Medien (und nicht etwa nur auf schwach polarisierbare) anwendbar. Wenn speziell ein Körper als aus einzelnen Atomen (im weitesten Sinne des Wortes) zusammengesetzt aufgefaßt werden kann und ihm eine (lokale) Clausius–Mossotti-artige dielektrische Funktion zugeschrieben wird, können alle relevanten Vielteilchen-van der Waals-Wechselwirkungen der beteiligten Materie in die Kraftberechnung einbezogen werden. Wie bereits bemerkt ist die Theorie sowohl auf Makro- als auch auf Mikroobjekte anwendbar. Die Kraft auf ein (lokal reagierendes) dielektrisches Mikroobjekt wird oftmals im Sinne einer Superposition von CP-Kräften auf einzelne, voneinander unabhängige Atome berechnet, zumindest im Falle schwach polarisierbarer Mikroobjekte. Durch den in dieser Arbeit vorgestellten Zugang kann der Einfluß sowohl der Gestalt des Mikroobjekts als auch der Vielteilchen-Wechselwirkungen auf die Kraft berücksichtigt werden, ohne dass eine Beschränkung auf schwach polarisierbares Material nötig ist. Wenn das Mikroobjekt auf nur ein Atom reduziert wird, erhält man genau die bekannte Formel für die CP-Kraft auf ein einzelnes Atom. Tatsächlich kann man aber nicht nur die Kraft auf ein isoliertes Atom bestimmen, sondern auch die Kraft auf ein Atom eines Mediums. Für ein solches ist die Kraft durch die Anwesenheit umliegender Atome des Mediums abgeschirmt, bei einem isolierten Atom tritt natürlich keine Abschirmung auf. Die grundlegenden Formeln gestatten es auch, die vdW-Wechselwirkung zwischen Atomen in der Anwesenheit von Körpern zu studieren.

Durch Spezialisierung auf den Fall planarer Strukturen konnten Formeln vom Lifschitz-Typ, die die Casimirkraft auf ebene Platten im Falle leerer Zwischenräume zwischen den Platten korrekt beschreiben, auch auf den Fall mit (lokal reagierendem) magnetodielektrischen Material gefüllter Zwischenräume verallgemeinert werden. Die mit dem Minkowski'schen Spannungstensor zusammenhängenden Probleme treten in diesem Fall sehr klar in Erscheinung. (Interessanterweise hatte sich Lifshitz in seinem grundlegenden Artikel [10] auf leere Zwischenräume beschränkt.) Für eine ebene, in ein Medium eingebettete Platte in einer Resonatoranordnung aus ebenen Platten erhält man im Grenzfall hoher Reflektivität auch die Verallgemeinerung von Casimirs bekannter Formel für zwei perfekt reflektierende Platten auf den Fall eines mit einem Medium gefüllten (anstelle eines leeren) Zwischenraumes zwischen den Platten. Wenn die Platte in ein Medium eingebettet

ist, kann sich die Kraft deutlich von der mittels des Minkowski'schen Spannungstensors berechneten unterscheiden.
Zusammenfassend kann gesagt werden, dass in dieser Arbeit ein allgemeingültiges theoretisches Fundament für die QED in beliebigen linear reagierenden Medien entworfen und im Detail ausgearbeitet wurde. Auf dieser Grundlage konnte eine einheitliche Theorie der Dispersionskräfte auf Objekte im Grundzustand (oder auch thermisch angeregte Objekte) entwickelt werden. Im Prinzip ist die Dispersionskraft auf ein makroskopisches Stück Materie ,,bloß" die (quantenmechanische) Lorentzkraft, die auf die konstituierenden Ladungen und Ströme wirkt, wobei selbige im Rahmen der makroskopischen Beschreibungsweise vollständig durch die zugrundegelegte lineare Materialgleichung spezifiziert sind. Wir sind der Meinung, dass dies ein praktisch von selbst einleuchtender und zufriedenstellender Standpunkt ist.

Ehrenwörtliche Erklärung

Ich erkläre hiermit ehrenwörtlich, dass ich die vorliegende Arbeit selbständig, ohne unzulässige Hilfe Dritter und ohne Benutzung anderer als der angegebenen Hilfsmittel und Literatur angefertigt habe. Die aus anderen Quellen direkt oder indirekt übernommenen Daten und Konzepte sind unter Angabe der Quelle gekennzeichnet.

Weitere Personen waren an der inhaltlich-materiellen Erstellung der vorliegenden Arbeit nicht beteiligt. Insbesondere habe ich hierfür nicht die entgeltliche Hilfe von Vermittlungs- bzw. Beratungsdiensten (Promotionsberater oder andere Personen) in Anspruch genommen. Niemand hat von mir unmittelbar oder mittelbar geldwerte Leistungen für Arbeiten erhalten, die im Zusammenhang mit dem Inhalt der vorgelegten Dissertation stehen.

Die Arbeit wurde bisher weder im In- noch im Ausland in gleicher oder ähnlicher Form einer anderen Prüfungsbehörde vorgelegt.

Die geltende Promotionsordnung der Physikalisch-Astronomischen Fakultät ist mir bekannt.

Ich versichere ehrenwörtlich, dass ich nach bestem Wissen die reine Wahrheit gesagt und nichts verschwiegen habe.

Jena, den 07. 02. 2008 Christian Raabe

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